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**HELIUM AND HEAVY WATER LEAKAGE FROM
A 3/8-INCH NEEDLE VALVE,
AS A FUNCTION OF
COMPONENT PRESSURE AND TEMPERATURE**

Report No 87-311-K

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ABSTRACT

The variance in helium and heavy water leakage with component pressure and temperature was observed for a 3/8-inch needle valve commonly employed in nuclear service. The experimental results were compared to theoretical predictions, and an attempt was made to explain some rather unusual leakage behaviour, such as self sealing of leaks, which was observed.

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16-281-451	819.312	February 1, 1988	87-311-K



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EXECUTIVE SUMMARY

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Heavy water leakage from nuclear components under typical or extreme operating conditions is routinely predicted by theory from observed D_2O leakage under different conditions, or from helium leak testing of that component. This study, undertaken at the request of the Nuclear Materials Management Department, is an effort to verify to what degree the real-life behaviour of components can be modelled by theory.

Helium and heavy water leakage testing was performed on a new 3/8-inch needle valve typical of those found at Ontario Hydro nuclear generating stations. The correlation between leakage, valve pressure, and temperature was established. (It should be noted that helium leakage for the valve, when closed properly, was $<1.6 \times 10^{10}$ atom cm^3/sec , the lower limit of detection. To obtain measurable leak rates so that correlations could be observed, the valve was "cracked" open very slightly.)

The helium leakage variation agreed reasonably well with theoretical predictions, with some small anomalies readily explained; eg by valve reseating. Heavy water leakage tests, however, revealed an interesting phenomenon--some kind of "self-sealing" effect. The D_2O leak rate would, typically, decrease by an order of magnitude within an hour of a valve pressure increase (after the initial increase in leakage). After several more hours, the leak rate would drop to a much lower level. Another valve of the same type also showed self-sealing effects, although less pronounced.

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An attempt was made to explain the self sealing phenomenon through microscopic, SEM, and x-ray analysis of the valve stem, and chemical analysis of the D_2O . These studies suggest that corrosion, with the resultant higher specific volume oxides; or residual deposits on the valve stem, containing elements also found in the heavy water, may be the mechanisms behind this phenomenon.

A comparison between helium and D_2O leakage of the same valve under identical conditions of temperature and pressure was also done. The observed D_2O leakage was only 1/90th of that theoretically predicted from the observed helium leakage, most likely due to the self-sealing effects.

It is evident, then, that theoretical leakage predictions for components of this type must be interpreted judiciously. It appears that relatively "normal" conditional changes such as those of temperature and pressure, or merely the passage of time, can cause distinctly non-ideal leakage behaviour.

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**HELIUM AND HEAVY WATER LEAKAGE FROM
A 3/8-INCH NEEDLE VALVE,
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R.E. Gnoyke

The variance in helium and heavy water leakage with component pressure and temperature was observed for a 3/8-inch needle valve commonly employed in nuclear service. The experimental results were compared to theoretical predictions, and an attempt was made to explain some rather unusual leakage behaviour which was observed.

INTRODUCTION

The heavy water leakage behaviour of a nuclear component under typical operating (or extreme) conditions is commonly predicted by theory from the results of helium leak testing of that component, or from observed D₂O leak rates under different pressure or temperature conditions/1/2/5/. The accuracy of such predictions depends on how closely the actual leakage behaviour of a component resembles that of an "ideal" leak. This study was undertaken at the request of the Nuclear Materials Management Department, to determine experimentally the variance of leakage of a typical component with temperature and pressure.

APPARATUS

The apparatus assembled to investigate the helium leakage characteristics of a Hoke 3/8-Gyrolok needle valve # 3812G6 is shown diagrammatically in Figure 1. A cylindrical oven chamber of stainless steel was flanged to the test port of a Veeco MS90 Mass Spectrometer Helium leak detector. This oven chamber would be under vacuum during leak testing. A thermostatically controlled band heater was clamped to the outside of the chamber.

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The 3/8-in Gyrolok valve to be tested has 3/8-in Gyrolok fittings at both the inlet and outlet end (see Figure 2). The inlet was swaged to a 3/8-in stainless steel tube which penetrates the lid of the chamber, and the outlet was left open. A thermocouple was attached to the outside of the test valve, as close as possible to the location of the valve seat. Valve temperature was then monitored with a Doric DS 300 thermocouple monitor, with a resolution of 1°C.

Helium pressure was supplied from a helium cylinder which was suitably valved into the system to allow slow, controlled filling and bleeding of the system, as shown. The system pressure was measured with one of two Ashcroft pressure gauges, 0-250 kPa (model S-30) and 0-10 MPa (Model S-1500).

Two calibrated helium leaks were available for leak detector calibration: a "slow" leak of 9.6×10^{-8} atm cm³/sec and a larger 3.2×10^{-4} atm cm³/sec leak.

The experimental setup for D₂O leakage testing is shown in Figure 3. The pressurizing system was the same as for He testing. The oven chamber was built into a closed loop circulation system, which included a Miran 101 infrared Spectrometer capable of measuring D₂O vapour in air in concentrations to 1000 ppm. The Miran has an integral fan (~20 L/min) which would move air from the chamber surrounding the valve to the spectrometer and back again. The vertical tube into the oven chamber (with the test valve on its end) was filled with liquid D₂O from the valve seat up to the swagelok fitting indicated on the diagram. When this was pressurized with helium, any leak in the valve would allow D₂O vapour into the chamber, (component temperature was >100°C), and the resultant concentration of D₂O vapour in air would be measured by the spectrometer. The calibration of the Miran varies somewhat with temperature; therefore, a water-cooled condenser was added to the loop to re-cool the circulating air to ambient temperature before it entered the spectrometer. A thermocouple sensor was installed at the Miran's inlet to monitor this temperature. A supply of dry nitrogen was available to flush out the loop between tests.

EXPERIMENTAL PROCEDURE

a) Preliminary Tests and Calibrations

These were performed to minimize experimental errors. They included the following:

- i) Calibration of the two pressure gauges in the filling system with a Mansfield and Green deadweight tester. Errors were noted and accounted for in subsequent calculations.
- ii) The Miran 101 infrared spectrometer was calibrated by injecting accurately known quantities of D₂O into a closed loop of known volume. Calibration factors were determined and incorporated in calculations.
- iii) The Veeco MS90 helium leak detector was calibrated using the two calibrated leaks. The sensitivity of the Veeco tended to vary somewhat from day to day; therefore, the instrument was calibrated every time an experiment was done.

- iv) There was some concern that the condenser which pre-cooled the loop air before it entered the Miran (D_2O leakage experiments) might be condensing out some of the D_2O vapour we were trying to measure. Calculations showed, however, that considering the starting humidity in the loop, and the temperature of the condenser, no vapour would condense out before a concentration of $\sim 14\,000$ ppm was reached. Since our tests were limited to a concentration of 1000 ppm (the maximum measurable by the Miran), this was not a concern. A simple test using a known leak rate for a measured interval confirmed this. No visible fogging of the condenser occurred until the concentration was well above 10 000 ppm.
- v) Since for D_2O leakage testing, the liquid D_2O in the valve was to be pressurized with helium, and some helium would undoubtedly dissolve in the D_2O , we had to be certain that the Miran infrared spectrometer would not respond to helium. The Miran measures the amount of absorption of infrared light at a wavelength corresponding to a strong absorption line for D_2O vapour. If helium gas also absorbed at this wavelength, the D_2O concentration measurements would not be accurate. To test this possibility, 100% helium gas was flushed through the spectrometer. The instrument indicated a " D_2O concentration" of 15 ppm. This very slight response could be ignored.

b) Procedure for Helium Leak Testing

The Gyrolok valve was mounted inside the oven chamber as shown in Figure 1. The valve could then be pressurized with helium as required. Initially, the valve and pressurizing system were purged of air by opening the valve and flowing helium through the whole system for some time. During leak testing, the oven chamber was evacuated by the leak detector's pumps, and helium leaking from the valve into the chamber, if any, would be detected. It should be noted that with this configuration, we measure the leakage across the seat and any leakage from the inlet Gyrolok swage fitting together. (The stem seal is downstream of the seat, and therefore is not pressurized.) The valve temperature could be varied as required by changing the oven chamber temperature.

A preliminary leak test of the valve was performed according to Ontario Hydro leak testing procedures/3/. In this case, the valve was mounted directly to the leak detector, and evacuated by it. A bag was placed around the valve, and helium was flushed through it. No leak of helium into the valve was observed. (The minimum detectable leak for the Veeco MS90 was 1.6×10^{-10} atm cm^3 /sec, as determined from calibrations.) Since the purpose of this study was to investigate variation of leakage with component temperature and pressure, a leaky valve was required. This was achieved by very slightly opening the valve until a leak was observed. (This meant that the leak we would be measuring would almost certainly be only across the valve seat.) The degree of opening the valve was varied for different experiments, choosing that amount which would result in leakages measurable over the full range of conditions for that particular experiment. The inlet valve of the helium leak detector was also throttled for some experiments, for the same reason. Leak detector calibrations for those experiments were, of course, done under the same throttled conditions.

Between experiments, the oven chamber was vented with air and re-evacuated several times to eliminate any residual helium in the chamber.

c) Procedure for D₂O Leakage Testing

For D₂O leakage tests, the valve was again "cracked" open slightly until a measurable leak rate was obtained. Because of the higher viscosity and surface tension of water (as opposed to helium) the degree of opening was larger than for the helium tests. The liquid D₂O (99.98%) in the valve and the 3/8-inch tube above it was pressurized with helium. The component temperature for these tests was kept above 100°C, since lower temperatures resulted in liquid leaks, and we would be measuring evaporation rates of the resultant drops of liquid D₂O. Water flows through the condenser jacket were adjusted so that air entering the Miran was at ambient temperature. The testing routine was as follows:

- The sampling loop (oven chamber, Miran, condenser, associated tubing) was flushed out with dry nitrogen. The loop was then "closed" again, and the concentration of D₂O vapour in it was monitored by the Miran. If there was a D₂O leak from the valve into the loop, the D₂O concentration would increase with time. The concentration versus time measurements were recorded for a suitably long period, and D₂O leak rates were calculated from them. Before the next experiment, the loop was again dried out with nitrogen. When the valve was to be left for longer periods of time, (eg overnight), room air was flushed through the chamber to keep it dry.

EXPERIMENTAL RESULTS AND DISCUSSION

a) Helium Leakage

The variation of helium leakage with $\Delta P (= P_{\text{valve}} - P_{\text{chamber}})$ for the artificially "leaky" 3/8-inch Gyrolok valve is shown, along with the associated error limits, in Figure 4. In order to compare these results with theoretical predictions, one must first establish what mode of gas flow exists for this leak, ie molecular, laminar, etc. The observed leakage variance with temperature and pressure suggest laminar flow. A detailed discussion of this determination is found in the Appendix. For laminar flow, Poiseuille's equation applies:

$$Q_{\text{gas}} = \frac{\pi}{8} \frac{r^4}{L} \frac{(P_1 - P_2) P_a}{\mu_{\text{gas}}}$$

Equation 1

where Q_{gas} = helium leak rate

r = radius of the leak channel

L = length of the leak channel

μ_{gas} = viscosity of helium

$P_1 - P_2$ = pressure differential between valve and oven chamber

$$P_a = \frac{P_1 - P_2}{2}$$

Reference 4

For a constant temperature, μ_{gas} is constant, and r and L are assumed to remain constant

$$\begin{aligned} \therefore Q_{\text{gas}} &= KP_a(P_1 - P_2) \\ &= K/2 (P_1 + P_2)(P_1 - P_2) \\ &= K_1(P_1^2 - P_2^2) \end{aligned}$$

$\therefore$ for laminar flow at a constant temperature, $Q_{\text{gas}} \propto P_1^2 - P_2^2$

- but in this case, $P_2 = 0$ (vacuum), $\therefore Q_{\text{gas}} \propto P_1^2 \equiv \Delta P^2$ or $Q_{\text{gas}} = K \Delta P^2$
($\Delta P = P_1 - P_2$ where $P_2 = 0$).

The value for K which gave the best fit to experimental data was determined by minimizing χ^2 . By varying K , (in the process of finding the minimum χ), we were, in effect, changing the theoretical dimensions of the leakage path, ie the r^4/L factor, until the most accurate predictions were obtained. $Q_{\text{gas}} = K\Delta P$ was then plotted (Figure 4) and a reasonable fit to experimental data was obtained. The average slope of the experimental leakage versus $K\Delta P$ curve seems to be slightly lower than that predicted by laminar theory. This trend is not easily explained - all the simple explanations lead to a steeper experimental curve. For example, if some leakage pathways were partially blocked by particulate matter or water in surface tension, one would expect these obstructions to clear with higher pressure, which would lead to a higher relative leakage at higher pressures. Or, if the valve reseated itself with pressure increases, (a phenomenon apparent in the D_2O leakage tests, later), it would almost certainly reseat in a "more open" position at the higher pressures, which would, again, cause a steeper experimental curve. Also, if any other leak(s) appeared as pressure increased, at the inlet swage fitting; for example, this would again cause a trend in the wrong direction. Considering all the possible variables, however, the data fits reasonably well to theoretical predictions.

A series of helium leakage versus ΔP experiments were done at several other temperatures (70, 120, 175, 225°C to observe the leak variance with temperature. The valve stem was not adjusted between these experiments, so this data should be directly comparable. The results are shown in Figure 5. Although the higher temperature runs (120, 175, 225°C) definitely had lower leakages than the cooler experiments, there is no clear trend with temperature. In fact, it appears as if some change in the valve has occurred to cause the large gap between those two groupings of data. Since the valve was being pressure-cycled for each run, and the temperature was changed between runs, a change in leak path, by re-seating of the valve stem, for example, would be a likely explanation. The Leakage Testing Handbook/4/ states that the shape of the leak path, and therefore, the magnitude of a gas (or liquid) leak may change with a variance in pressure or temperature. It goes on to say that pressure increases may open up new pathways, resulting in higher than predicted flow rates; and that temperature changes are even less predictable, with an increase or a decrease in temperature changing the leakage path.

In this case, the leakage path appears to have changed between experiments, probably when the pressure was cycled down from 10 MPa at the end of one experiment to 100 KPa at the beginning of the next. No anomaly was observed during any one experiment, ie no sudden "jumps" in the curve. A change in leak detector sensitivity between runs can be ruled out, since calibrations were done after each run. In any case, we are not able to gain much leakage versus temperature information from the data plotted in Figure 5.

It was suspected that the biggest contributor to leak path changes was the pressure cycling inherent in the nature of this last series of tests. To reduce the number of pressure changes, it was decided to approach the leakage versus temperature investigation as follows: keep the pressure constant, and vary the temperature through the desired range. Then, the pressure would be increased, and the whole temperature cycle repeated. The temperature was varied slowly, and leakage measurements were done with both increasing and decreasing temperatures, and the results averaged, to counteract any hysteresis effects. The experiments were done at three pressures, 1.0, 4.9 and 9.8 MPa. The result of the 4.9 MPa experiment, and the corresponding theoretical laminar flow curve, is shown in Figure 6. As can be seen, the experimental leakage follows laminar theory very well. During the 1.0 MPa experiment, however, there appear to be leakage path changes, see Figure 7. One half of the experimental curve is well above laminar theory, and the other half is well below. If we assume that the sudden drop in leak rate is due to a change in leak path because of the temperature variation, then the first part of the experimental curve should still follow laminar theory, and the part after the change should again follow laminar theory, although for a different K in the equation

$$Q_{gas} = \frac{K}{\mu_{gas}}$$

since the leak dimensions would have changed. When the theory was fitted to the two halves of the curve separately, this does appear to be true (Figure 8). Similar behaviour was observed by J. Marr for a gasket leak/6/ (Figure 9), although in his case, the leak geometry changes were caused by pressure changes. The 9.8 MPa experiment fits reasonably well to laminar theory (Figure 10), although it appears there may have been some geometry changes of smaller magnitude than for the 1.0 MPa experiment.

b) D₂O Leakage

Poiseuille's equation for laminar liquid flow is:

$$Q_{liq} = \frac{\pi}{8} \frac{r^4}{L} \frac{(P_1 - P_2)}{\mu_{liq}}$$

Equation 2

where Q_{liq} = D₂O leak rate

μ_{liq} = viscosity of D₂O

all others - same as for gas leakage/4/7/.

For a constant temperature, μ_{liq} is a constant, and we assume that the dimensions of the leak remain the same. The D_2O leakage should then be proportional to ΔP , ie $Q_{\text{liq}} = K(P_1 - P_2) = K \Delta P$.

To confirm the above, an experiment was done at a component temperature of 120°C , with ΔP varied between 2.4 and 9.7 MPa. The observed leakage was not at all as predicted. It increased with pressure at a higher rate than if it were proportional to ΔP between 2.4 and 7.3 MPa, and then there was virtually no increase between 7.3 and 9.7 MPa. It was decided to repeat the experiment under exactly the same conditions. This time, not only was the leak rate not proportional to ΔP , but the leak rates had decreased dramatically from those measured in the first instance. The results of these two experiments are shown in Figure 11. Obviously, leak path changes had occurred. Because of the erratic nature of these curves, it appears as if the changes were taking place throughout the duration of the experiments. It was decided to observe the leakage under truly constant conditions. The valve was "cracked" open just a little more, since the leakage had become quite small. The pressure was then increased from 100 kPa*¹ and then, for a period of thirty minutes, both ΔP and temperature (120°C) were kept constant. The D_2O leakage was measured every 10 minutes for the duration of the experiment. During this time, the leak rate decreased by approximately 1/3 of its initial magnitude (Figure 12). Therefore, even under constant conditions of pressure and temperature, the D_2O leak rate was decreasing with time.

Since a D_2O leakage versus ΔP study was out of the question under these conditions, it was decided to investigate the leakage versus time phenomenon in greater detail.

After some time at 100 kPa and 120°C , the valve was re-pressurized to 2.4 MPa and the D_2O leakage was measured periodically over thirty hours, to observe longer term effects. The results of this experiment are shown in Figure 13. The leak rate increases dramatically (factor of 5) in the first hour after repressuring, then decreases quite quickly again for approximately four hours. After that, the leak rate decreases at a slower rate. One possible explanation for this behaviour is as follows: some of the leakage channels may have become blocked, for some reason, while the valve was at 100 kPa and 120°C . If there is only one leakage path, (unlikely), it may have become partially blocked. When the pressure was increased to 2.4 MPa at the beginning of the test, the passages were cleared somewhat by the higher pressure and continued to clear for ~1 hour; therefore, the sudden increase in leak rate. Then, under constant temperature and pressure, whatever caused the blockage in the first place began to reform again, and the leak rate decreased. A likely blocking mechanism is inherent in the nature of the leakage itself, that is, liquid D_2O at 120°C and under pressure is forced through some kind of leak channel(s) towards the outside of the valve, where the pressure is at ambient. Somewhere within the channel, the pressure is low enough so that the D_2O boils off and travels the rest of the way out as water vapour. At that point, it seems quite likely that some kind of deposit (due to solutes in the D_2O) could be left behind to partially (or totally) block the channel(s). Also, if the steam is causing any oxidization of the stem or seat material (SS316), however minute, the higher specific volume of the oxide film/6/ would cause a

1. *Note: The "base" pressure of 100 kPa was necessary to keep the liquid D_2O in the valve from boiling at 120°C .

shrinkage of the leakage path. This film, if not well attached, could be "washed out" during a positive pressure change, only to reform again, perhaps at a different location along the channel, once a steady pressure state exists. In any case, we have observed some type of self-sealing effect.

The experiment was repeated, with the results shown in Figure 14. For comparison, the previous experiment is also shown on this graph. It appears that once the blockage has occurred (at 2.4 MPa), reducing the pressure did not disturb it, as could be expected. When the valve was repressurized, the leakage started out where it had been at the end of the previous test. Some degree of clearing seems to have taken place, perhaps due to a reseating of the valve stem with the ΔP increase, and then the "self-sealing" effect took over. The leakage at 30 hours was approximately the same as for the previous experiment. The next logical test, then, was to increase the pressure further, to see if the same clearing-sealing cycle would take place.

The component pressure was increased from 2.4 MPa to 4.8 MPa, and the leakage was measured periodically for thirty hours. Then, the pressure was again increased, to 7.3 MPa, and after thirty hours, to 9.7 MPa. The measured leak rates for these tests, as well as the 100 kPa $\rightarrow$ 2.4 MPa test, are shown in Figure 15. In every case, the leak rate increased very quickly with the pressure increase, then decreased again, as for the earlier experiment. Both the increases and subsequent decreases appeared to occur at a faster rate than for the 100 kPa $\rightarrow$ 2.4 MPa experiment. In the case of the 7.3 $\rightarrow$ 9.7 MPa increase, the leakage was so high that the condenser fogged for the first two minutes, even with very fast dry nitrogen flushing. Within four minutes, however, the leak rate had decreased to the point that it was measurable, and continued to decrease to the same range as the others. After 24 hours, the leakage versus time elapsed curves had levelled off. The leakages at their respective ΔP were compared at that point, but no trend evolved.

The system was left at 9.7 MPa for an extra long test, and after four days, the leak rate had decreased further, to 1.22×10^{-2} gm/day (from 4.3×10^{-2} gm/day at 24 hours). Therefore, what we have observed is some kind of self-sealing effect that appears to continue over long periods of time. Presumably, it may go on until the leakage has totally stopped. This particularly makes sense if, as described in the earlier hypothesis, the sealing effect is somehow due to the leakage itself.

After the valve had been left at 120°C, 9.7 MPa for four days, and the leakage was decreasing with time quite slowly, leakage versus ΔP measurements were done with decreasing ΔP , going on the assumption that the blockage would not be disturbed in this manner. This indeed appeared to be true, since the measured D₂O leak rates follow theory very well (Figure 16). Therefore, as observed for helium leakage, the leakages would behave in a predictable manner, were it not for all the other "reseating" or "self-sealing" effects, etc.

It was decided to increase the pressure again back to 9.7 MPa, after the measurements with decreasing ΔP were complete. The leakage again increased, from the 1.2×10^{-2} g/day that had been observed after four days at 9.7 MPa, to 4.2×10^{-2} g/day. Within 30 hours, however, the leak had "sealed" itself again, down to 1.0×10^{-2} g/day. It appears, then, that if this valve had been under steady pressure and temperature conditions for quite a long time, (a day, at least), and the leak rate is known; then with a downward

pressure change, the new leak rate could be predicted by laminar liquid theory. An upward pressure fluctuation, however, would result in leakages much higher than predicted, at least initially.

To gain more insight into the nature of the self-sealing effect, three studies were undertaken: a microscopic analysis of the valve stem (both optical and scanning electron microscope), a quantitative x-ray analysis of the stem to identify any foreign matter on its surface, and a chemical analysis of the D_2O .

Optical micrographs (mag x 18) of the valve stem in the area of contact with the valve body are shown in Figure 17. Also shown for comparison is a "virgin" stem. A sharp line can be seen approximately half way along the conical part of the stem, where the metal was deformed slightly. This was most likely where the stem contacted the valve body. This then is the actual "sealing" line. Upstream of this line, ie on the high pressure side of the seal, a "rusty" patch is observed in one orientation of the stem, and not apparent on the other. On the downstream (low pressure) side of the seal, a circumferential ring of "rust" and brownish residue is visible. This ring appears to have some topography to it, as seen in the low magnification (x 22) scanning electron microscope (SEM) photograph (Figure 18). Also observed in the vicinity of this ring were two whitish deposits, one of which is shown in Figure 19. A diagram of the valve stem and seat and the relative positions of these observed features is shown in Figure 20.

The degree of "rusting" in the SS316 valve stem after approximately one month of testing was surprising. It certainly lends support to the theory of oxidization of the valve materials, with the resultant higher specific volume oxides, being a potential self-sealing mechanism. The thickness of the downstream ring of residue and the whitish deposits also go along with the other hypothesis discussed earlier: that the water boiling off may leave residue behind, which could block leakage pathways. Perhaps the observed ring is such residue, which had been "washed out" during pressure increases.

One possible type of leakage path can be seen in Figure 21. This photograph was taken on the "sealing line", and shows what appears to be mechanical damage/9/, probably from metal-to-metal binding during opening and closing of the valve.

The results of energy-dispersive x-ray analysis (EDXA) of the valve stem are shown in Figure 22. The elemental composition of 316 SS/10/ is also shown, and a "virgin" stem was analyzed for comparison. The most noticeable difference between the virgin stem and the "rusty" area of the used stem is the presence of chlorine in the latter, and not in the former. The chlorine probably originated in the D_2O ; subsequent chemical analysis of the D_2O (Figure 23) found traces of Cl ($5 \mu\text{g/l}$)*². Note also that the D_2O was slightly acidic with a pH of 6.3.

2. *It should be noted that oxides are not observed with the EDXA equipment which was used; sodium is the lightest element detectable. Also, this technique will, in general, analyze the specimen to a depth of approximately $3 \mu\text{m}$ down from the surface. In the case of a thin coating, then some of the composition of the base material will "show through".

The white deposit appeared to be composed mainly of manganese - 50%, sulphur - 28% and chlorine - 12%, with traces of potassium, copper and zinc also appearing. The EDXA overlaps the sulphur and molybdenum peaks, and Mo usually appears as S in the analysis. Since the chemical D_2O analysis showed only a small presence of sulphates ($4 \mu\text{g/l}$), perhaps the high "S" peak was really due to Mo. Lubricating grease on the threaded part of the stem may be the source; a sloppy application during manufacture may have resulted in some grease on the lower stem. Manganese concentration in the D_2O was $<20 \mu\text{g/l}$ (the lower limit of detection). Any further comment on the origins or the significance of the presence of these elements on the stem as related to the observed leakage phenomena is beyond the scope of this report.

An attempt was made to duplicate the self-sealing phenomenon in another valve. A second valve, identical to the first, was mounted in the oven chamber, and D_2O leakage tests were performed. The results are shown in figures 23 and 24. The observed D_2O leakage behaviour was somewhat different than that for the first valve. In the 2.4 MPa case, (up from 100 kPa at $T = 0$), the leakage decreased more or less linearly over the 5-day measurement period to approximately 1/7th of that at the beginning of the test. This is in direct contrast with the behaviour of the first valve, where most of the leakage decrease would occur in a matter of hours, and only smaller, gradual decreases were observed after that.

When the pressure was increased to 4.8 MPa, the leak rate started out at approximately twice what it was at the end of the 2.4 MPa, as one would predict with the (linear) theory. For two hours the leakage seemed to drop slightly, but after a day, the leak rate had increased, seemingly spontaneously. We had observed leakage increases with the other valve, but always in conjunction with a change in either ΔP or temperature. In this case, there was no such change.

The pressure was increased to 7.3 MPa, and an 8-day experiment was undertaken (Figure 24). Over this long period, ΔP dropped slowly (because of the leakage), and the pressure was "topped up" twice. The pressure history is also shown in Figure 24. It should be noted that the leak rate values plotted had been linearly corrected for pressure, eg if the leak rate was measured at $\Delta P = 7.2 \text{ MPa}$, it was corrected to what it would have been at 7.3 MPa, according to $Q_{\text{liq}} \propto \Delta P$.

The leakage decreased for an hour and a half after the pressure increase to 7.3 MPa, but then suddenly increased (at #1 on the graph), with a slow decrease in ΔP of only 0.1 MPa. With the other valve, leakage did not seem to vary unpredictably with ΔP decreases, even large ones.

The leakage then fluctuated through a narrower range for 6-1/2 hours, after which it dropped. Then, at #2 on the graph, it decided to jump up again, with only a gradual ΔP decrease, as before. At #3, a 10% upward adjustment in ΔP was made, resulting in a 5-fold increase in leak rate 70 hours later. The leak rate then went down to approximately where it was before. Another upward pressure adjustment started the leak rate increasing again, at #4.

It is difficult to explain the differences in leakage behaviour of the two valves under similar conditions. The leak rates at the beginning of the two experiments were similar,

therefore, the effective dimensions of the leakage path(s) must be similar. Perhaps there were fewer, but relatively larger leakage paths for the second valve, a condition which would result in leakages in the same range; but the larger paths might be more easily washed free of blockage without major ΔP or temperature changes. Of course, the shape of the channels, the smoothness of the walls, or any number of variables might also affect the leakage versus time behaviour. It should be noted that whenever an increase in leak rate occurred, whether spontaneous or not, a decrease would eventually follow. There does appear to be some kind of self-sealing mechanism at work here, although it seems to operate on a different time scale than that observed before.

A microscopic inspection of the valve stem after this series of test revealed an appearance similar to that of the first valve. There were "rust" spots upstream and downstream of the valve-seat line, although not as much as in the first valve. This is to be expected, since the duration of this test was considerably shorter. A ring of brownish residue was also observed downstream of the seal, as before. No whitish deposits were seen.

CORRELATION OF HELIUM LEAKAGE TO D₂O LEAKAGE

The helium and heavy water leakages were measured under identical conditions of temperature and pressure in order to find the relationship between the two. The temperature in both cases was 150°C, and ΔP was 1.1 MPa. (In the case of He leakage, gauge pressure was only 1.0 MPa, but the 0.1 MPa from ambient to vacuum was added.) The D₂O leakage was measured immediately after. The observed leak rates were as follows:

$$\text{Helium leak rate} \quad \equiv Q_g = 2.67 \times 10^{-3} \text{ atm cm}^3/\text{sec}$$

$$\text{D}_2\text{O leak rate} \quad \equiv Q_L = 5.04 \times 10^{-1} \text{ g/day}$$

If we assume that the flow regime was laminar in both cases, and since the temperature and ΔP conditions were identical, then we do not need to know the dimensions of the leakage path(s) in order to predict one leakage (Q_L) from the other (Q_g). By dividing equation 1 by equation 2, we obtain the relationship:

$$Q_L = \frac{\mu_g}{\mu_L} \frac{Q_g}{P_a},$$

Equation 3

which is independent of leak geometry. In this case,

$$Q_g = 2.67 \times 10^3 \text{ atm cm}^3/\text{sec}$$

$$P_a = \frac{1.1 \text{ MPa} - 0}{2} = 0.55 \text{ MPa} = 5.43 \text{ atms}$$

$$\mu_g \equiv \mu_{\text{He}} @ 150^\circ\text{C} = 2.47 \times 10^{-4} \text{ poises /7/}$$

$$\mu_L \equiv \mu_{\text{D}_2\text{O}_{\text{liq}}} @ 150^\circ\text{C} = 2.16 \times 10^{-3} \text{ poises.}$$

The μ_L value was calculated from a semi-empirical formula put forth by Khan/2/ which predicts the viscosity of liquid heavy water between 25 and 300°C.

From Equation 3, then, we obtain a predicted D₂O leak rate of 5.63×10^{-4} cm³/sec, which in the D₂O leak rate units is 4.59×10^1 g/day. Therefore, the observed D₂O leak rate (0.5 g/day) is 1/90th of that predicted from observed helium leakage by laminar theory (45.9 g/day).

There are several aspects of this experiments and the above correlation which may explain why the observed D₂O leakage is smaller than predicted.

We have assumed that the dimensions of the leakage channel(s) have remained constant throughout. In reality, however, the effective diameter of the channel(s) would be reduced by an immobile layer of liquid next to the channel wall/4/.

Equation 3 predicts liquid flow, when, in fact, we have liquid flow to a point in the flow channel, and vapour flow for the remainder. The D₂O vapour at that point would exert a back pressure on the liquid in the channel, equal to the saturation vapour pressure at the prevailing temperature. At 150°C, the saturated vapour pressure of water is quite high, 4.69 atms/7/. This means that, if we assume that the vapour would flow more easily through the channel than the liquid, and therefore, that the liquid flow rate would determine the total mass flow rate through the channel, then the ΔP for the liquid is only $5.43 - 4.69$ atms = 0.74 atms. Since liquid laminar flow is proportional to ΔP , then the predicted D₂O leakage would drop to

$$\frac{0.74}{5.43} \times 4.59 \times 10^1 = 6.26 \text{ g/day ,}$$

ie, only twelve times the observed flow rate. It should be emphasized that this is an extremely simplistic view of the problem, described here only to show that there may be an effect. For one thing, our assumption that the liquid vapourized within the channel means that the length of the channel, as far as the liquid is concerned, has decreased, leading to higher flows. Burrows/8/ describes a detailed treatment of this problem which takes into account the surface tension of the liquid, capillary dimensions, tube-end effects, water-to-wall contact angles, gravity and head of water, etc. Since we do not even know the physical dimensions or shape of our leak, this technique was not attempted.

Lastly, we have totally ignored the possible effects of "our" self-sealing mechanisms for D₂O leaks. The test was done as quickly as possible to reduce this effect, but there was almost certainly some contribution towards lower than predicted liquid leakages.

CONCLUSIONS

Apparatus has been constructed to measure the helium and heavy water leakage of a 3/8 Gyrolok needle valve under various temperature and pressure conditions. The relationships between leakage and component pressure and temperature have been observed, and compared to those predicted by theory. Although the leakage versus component pressure and leakage versus component temperature curves agree reasonably well with theory in the best instances, many unpredictable fluctuations in leakage were observed. It was attempted to explain these discrepancies. An unexpected "self-sealing" effect was observed in the case of D₂O leakage. An attempt to explain this phenomenon has been made, through microscopic, SEM, and x-ray analysis of the

valve stem, and chemical analysis of the D_2O . Deposits on the valve stem (which are suspected of causing the "self-sealing" effect) showed constituent elements not present on an unused stem. These same elements were found in the heavy water, although in relatively small concentrations. Helium and D_2O leakages were measured for the same component under identical conditions and the results were compared to theoretical predictions. The D_2O leakage predicted from the observed He leak rate was found to be higher.

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APPENDIX I

Determination of Flow Regime

In order to compare the experimental Q_{gas} versus ΔP or T curves to theoretical predictions, one must establish the flow regime which exists in this case. The Leakage Testing Handbook/4/ states that it is possible to predict the mode of flow through a leak channel from a knowledge of the rate at which gas passes through a leak under a differential pressure in the order of one atmosphere.

<u>Mode</u>	<u>Leakage Rate (atm cm³/sec)</u>
Turbulent	$>10^{-2}$
Laminar	10^{-1} to 10^{-6}
Transition	10^{-4} to 10^{-7}
Molecular	$<10^{-7}$

In this case, at 22°C and $\Delta P = 1$ atm, the helium leak rate is $\sim 1 \times 10^{-4}$ atm cm³/sec (Figure 4). This would suggest laminar flow. The ranges overlap, however, and this leak rate is also at the upper limit for transitional flow. Examination of the leakage versus temperature characteristics tends to eliminate this possibility, since:

For Laminar Flow

$$Q_{gas} \propto \frac{1}{\mu_{gas}} \text{ where } Q_{gas} = \text{leak rate}$$

μ_{gas} = viscosity of the gas/4/5/.

For helium, viscosity increases with temperature, and therefore, Q_{gas} would decrease with temperature.

For Molecular Flow

$$Q_{gas} \propto \sqrt{T}$$

References 4 and 5

Therefore, Q_{gas} would increase with temperature.

For Transitional Flow, The leak rate would behave as a combination of the two above.

In our case, the leak rate decreases with temperature, possibly even at a higher rate than that predicted by laminar flow theory. For transitional flow, it should decrease at a slower rate. This suggests, again, laminar flow.

It should also be noted that there are, of course, other, more numerical methods to predict flow regime, eg using Reynolds numbers/4/5/, but these rely on assigning dimensions to the leakage path, which in itself would be guesswork in this case.

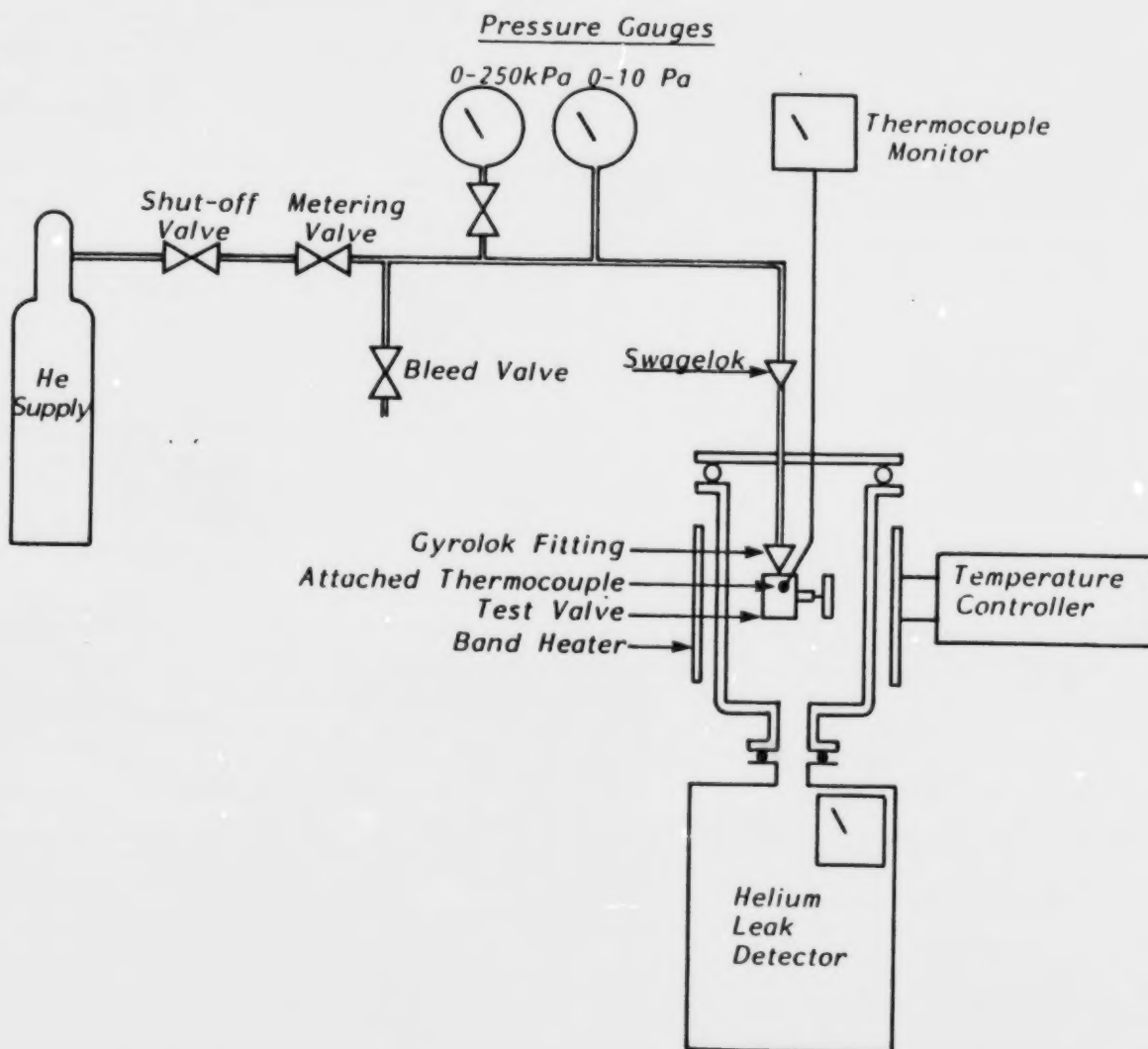


FIGURE 1
EXPERIMENTAL SET-UP - HELIUM LEAKAGE TESTS

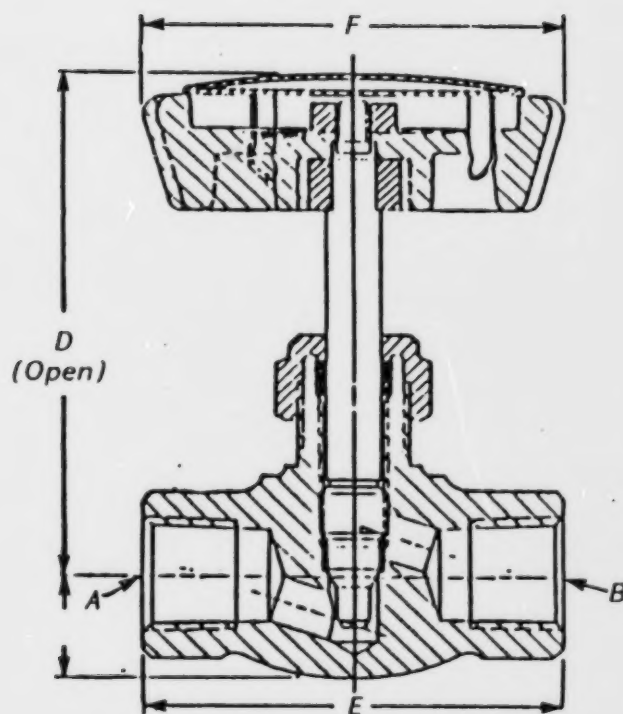


FIGURE 2
3/8" HOKE GYROLOK NEEDLE VALVE #3812G6

A = Inlet = 3/8" Gyrolok
B = Outlet = 3/8" Gyrolok
D = 70mm E = 65mm
F = 48mm H = 13mm

Materials: Stem 316SS Body 316SS
Packing: Teflon

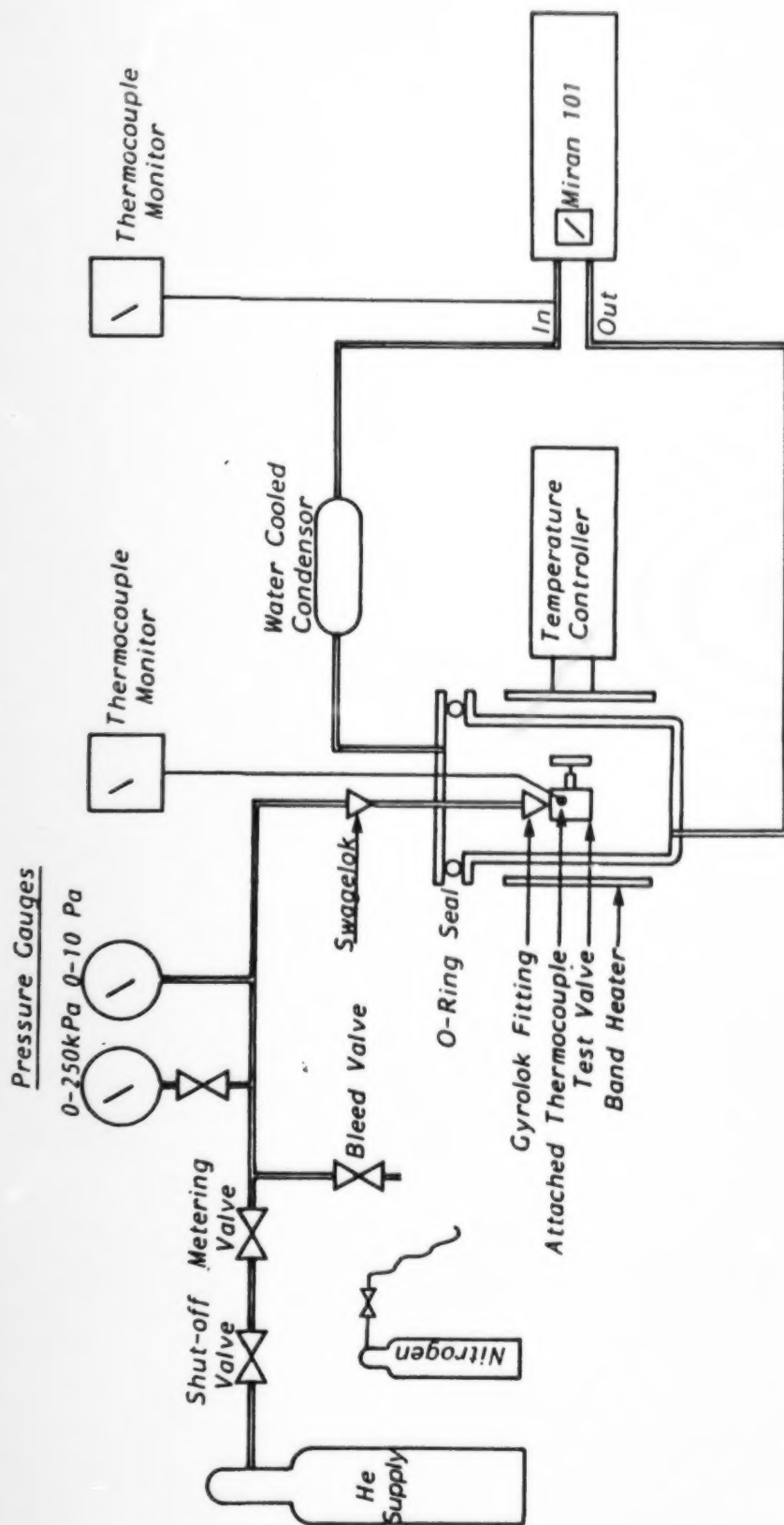


FIGURE 3
EXPERIMENTAL SET-UP - D₂O LEAKAGE TESTS

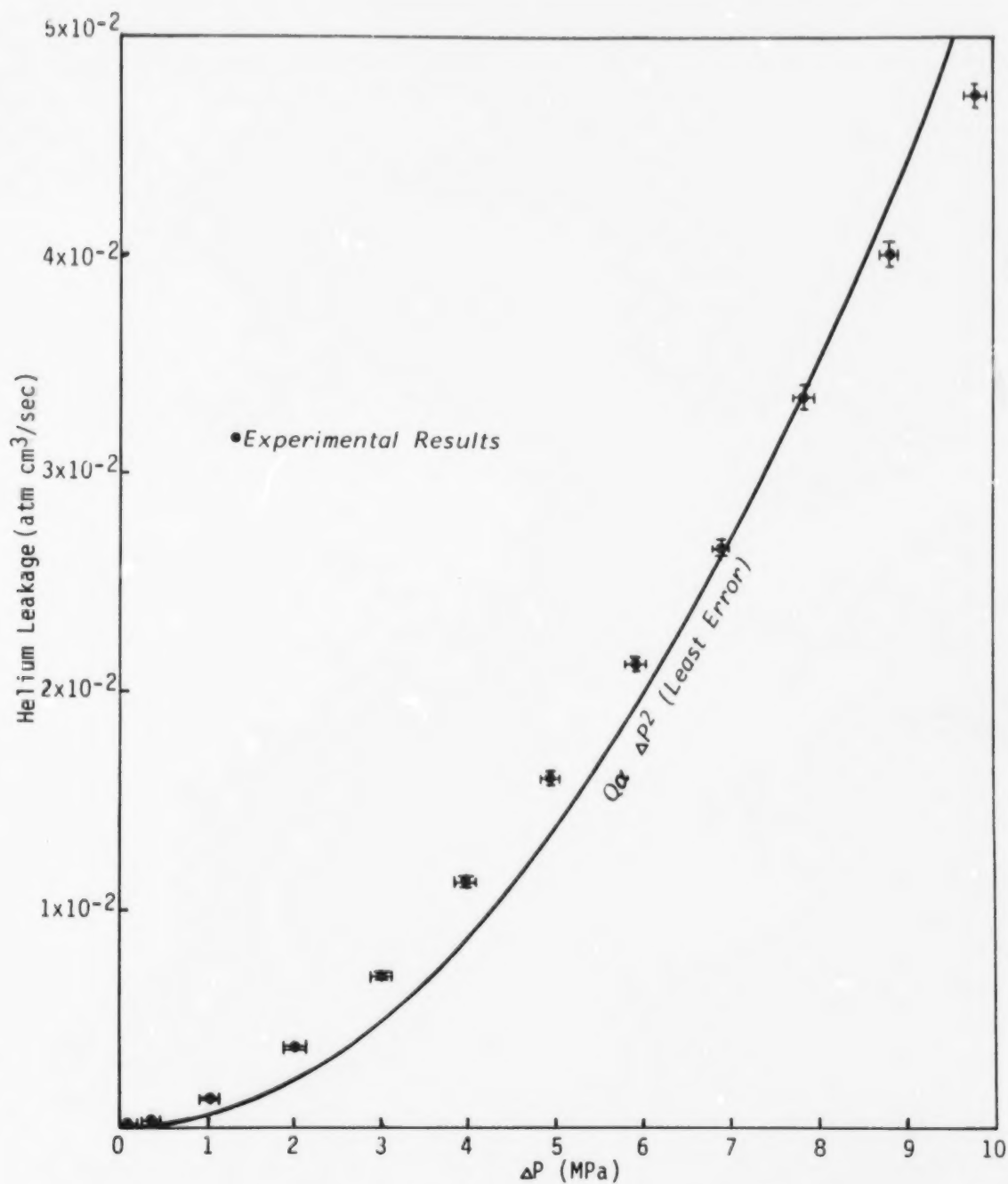


FIGURE 4
 HELIUM LEAKAGE VS ΔP @22°C
 3/8 GYROLOK VALVE

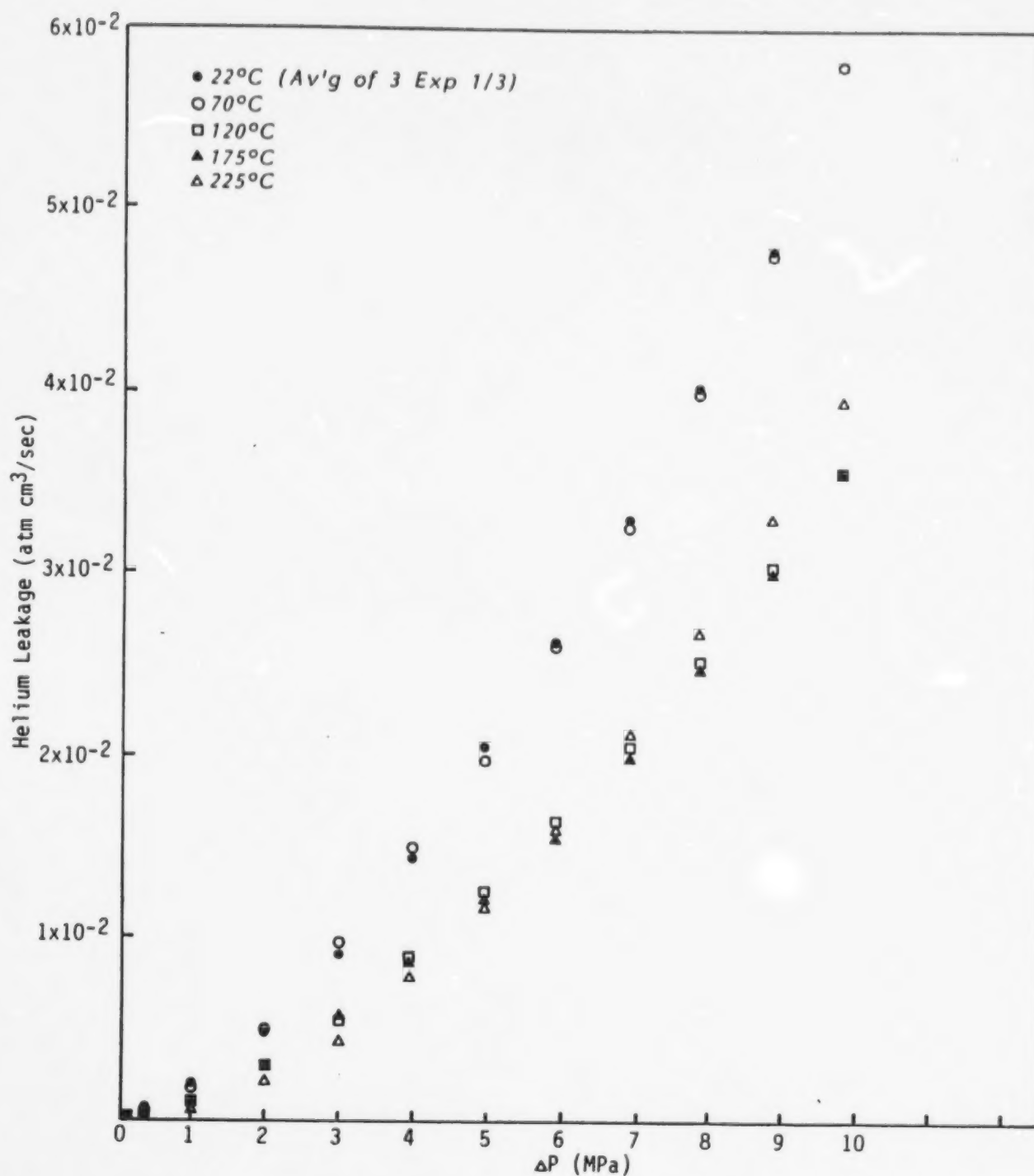


FIGURE 5
HELIUM LEAKAGE VS ΔP AT VARIOUS TEMPERATURES
3/8 GYROLOK VALVE

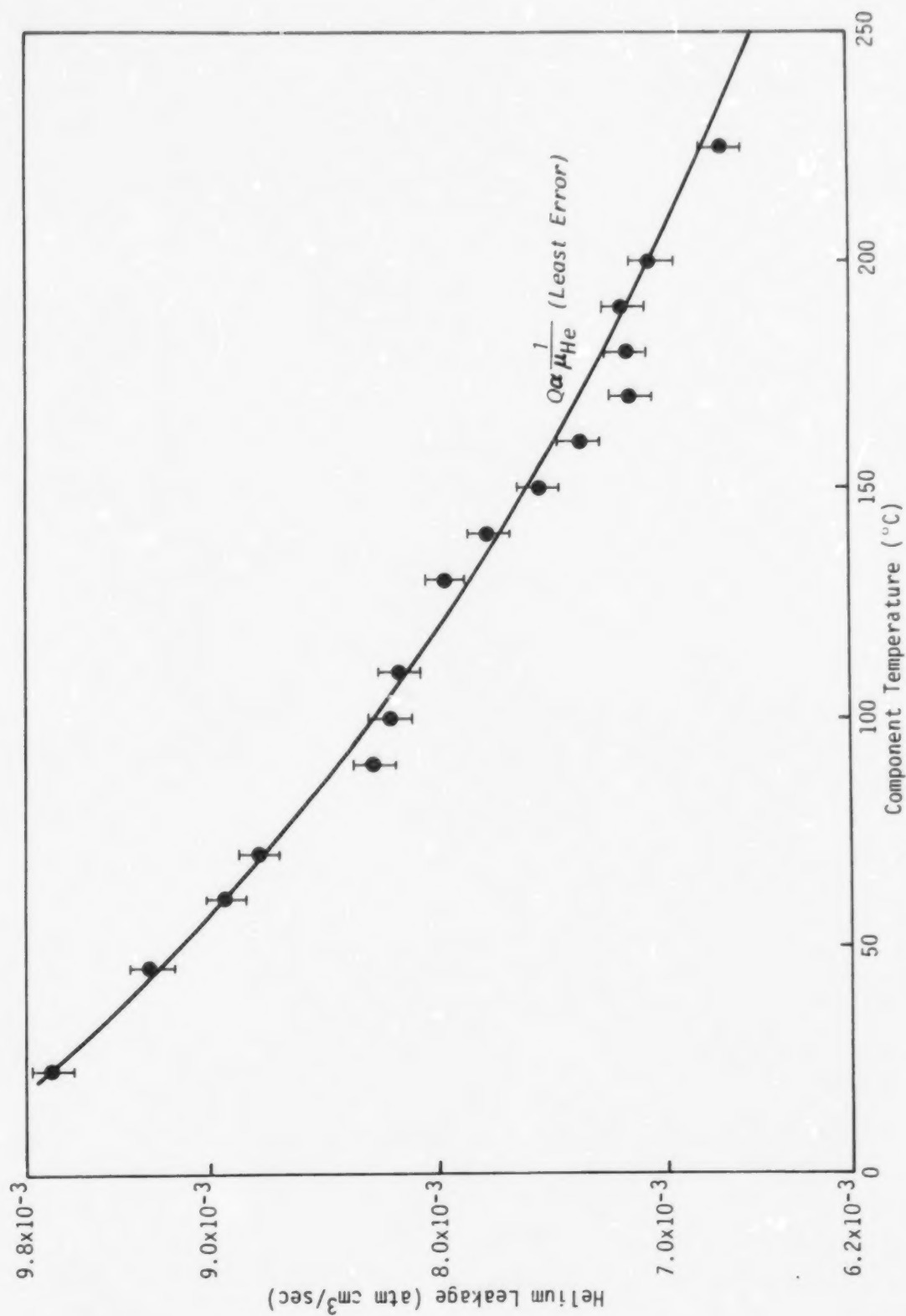


FIGURE 6
 HELIUM LEAKAGE VS COMPONENT TEMP.
 - 3/8" Gyrolok Needle Valve #3812G6
 - ΔP Across Seat = 4.9 MPa

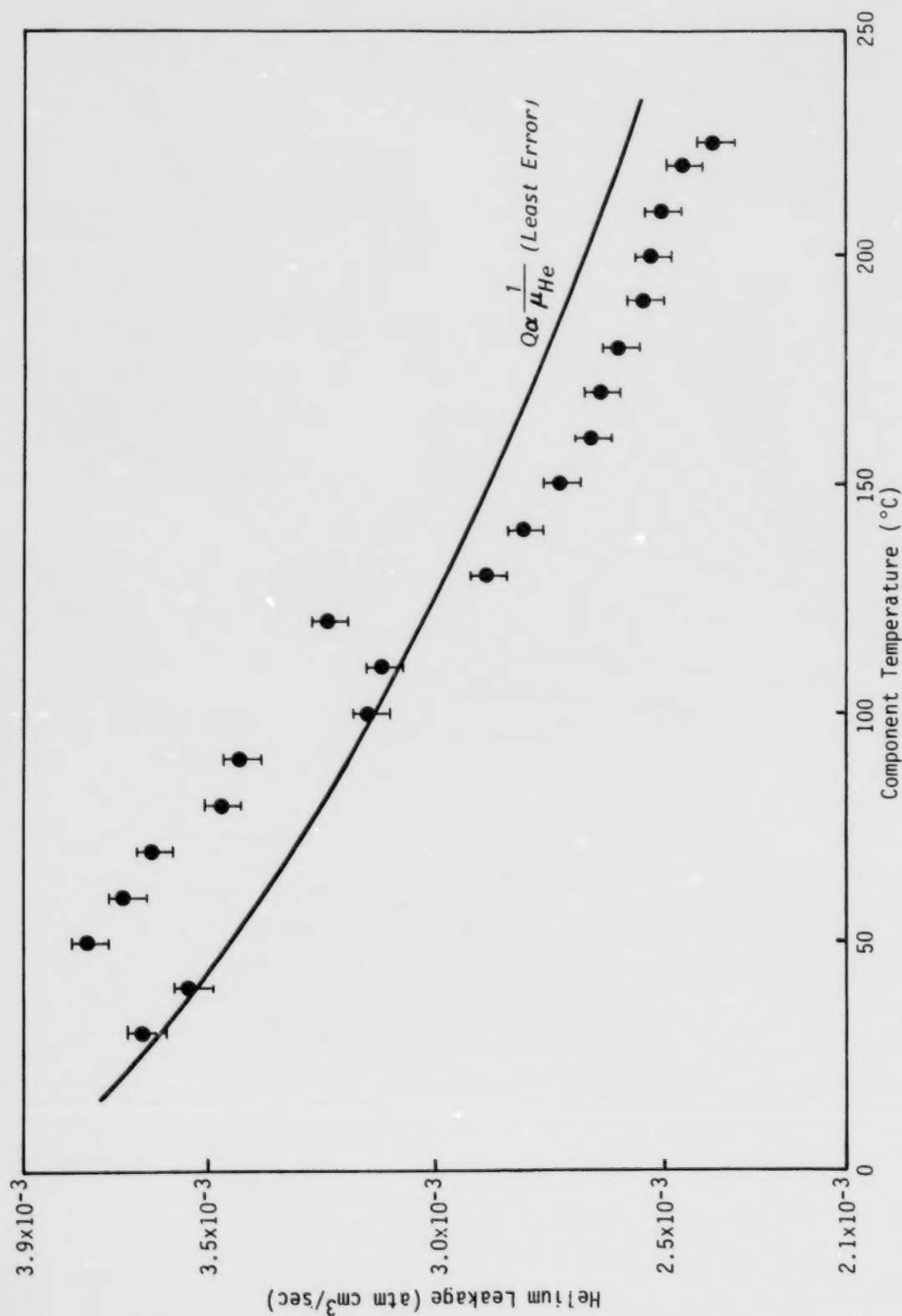


FIGURE 7
 HELIUM LEAKAGE VS COMPONENT TEMP.
 -3/8 Gyrolok Valve#3812G6
 -ΔP Across Seat 1.0 MPa

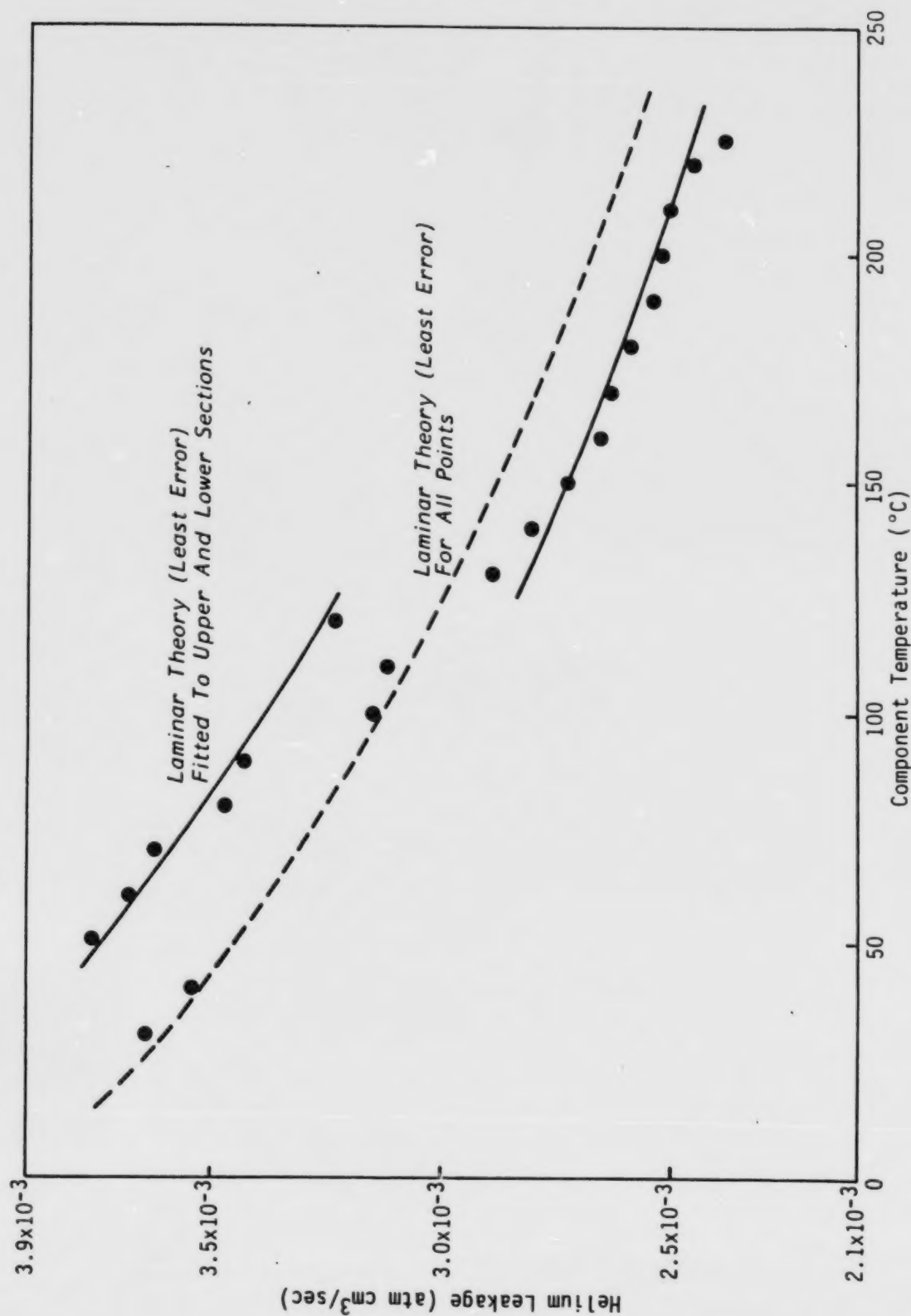


FIGURE 8
HELIUM LEAKAGE VS COMPONENT TEMP.
-3/8 Gyrolok Valve# 3812G6
-ΔP Across Seat 1.0 MPa

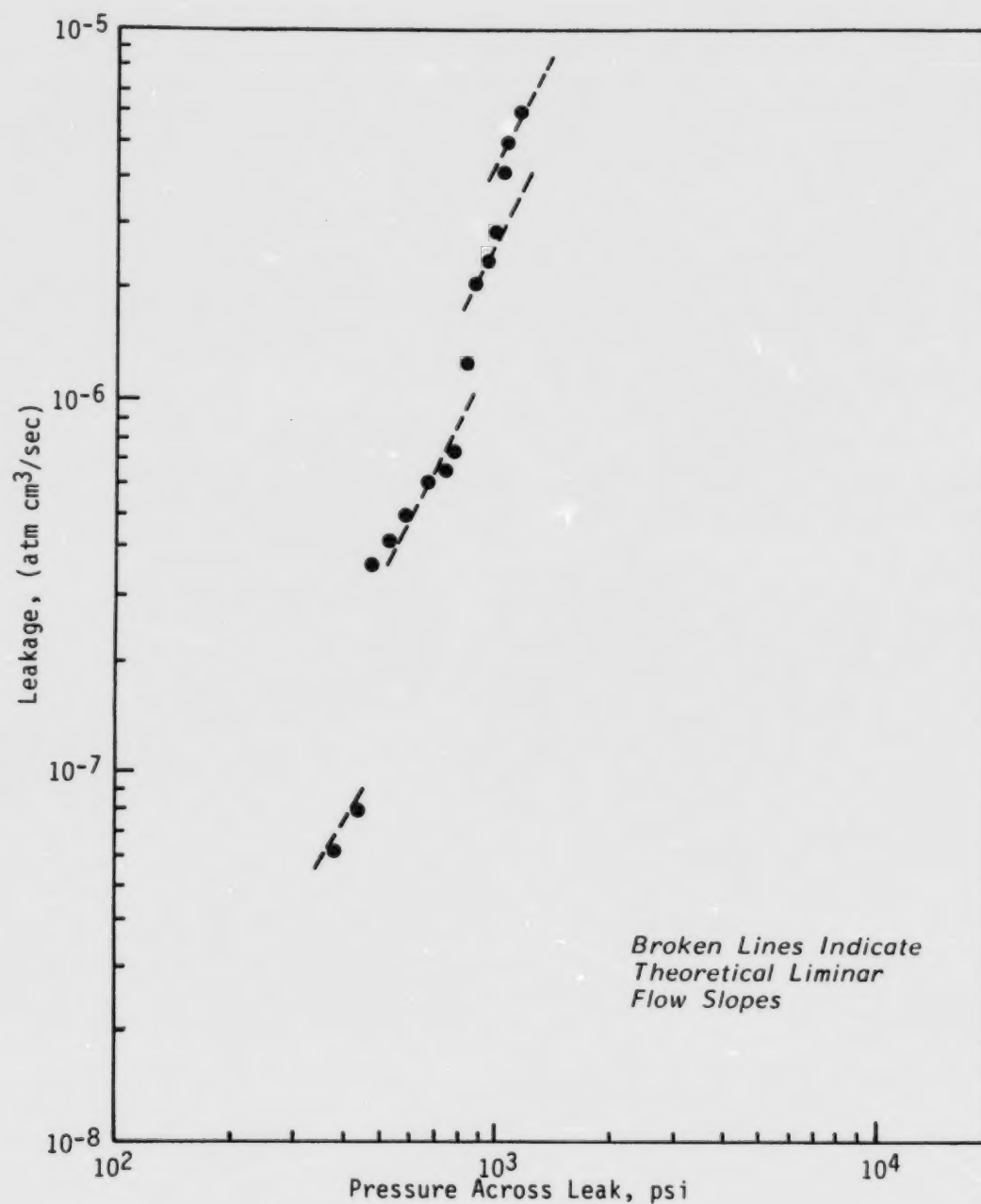


FIGURE 9
GEOMETRY CHANGE IN A GASKET LEAK
(Redrawn from J.W. Marr, Study of Dynamic and
Static Seals for Liquid Rocket Engines, Final Report
Phase II, Contract NAS 7-102 (Ref 6).

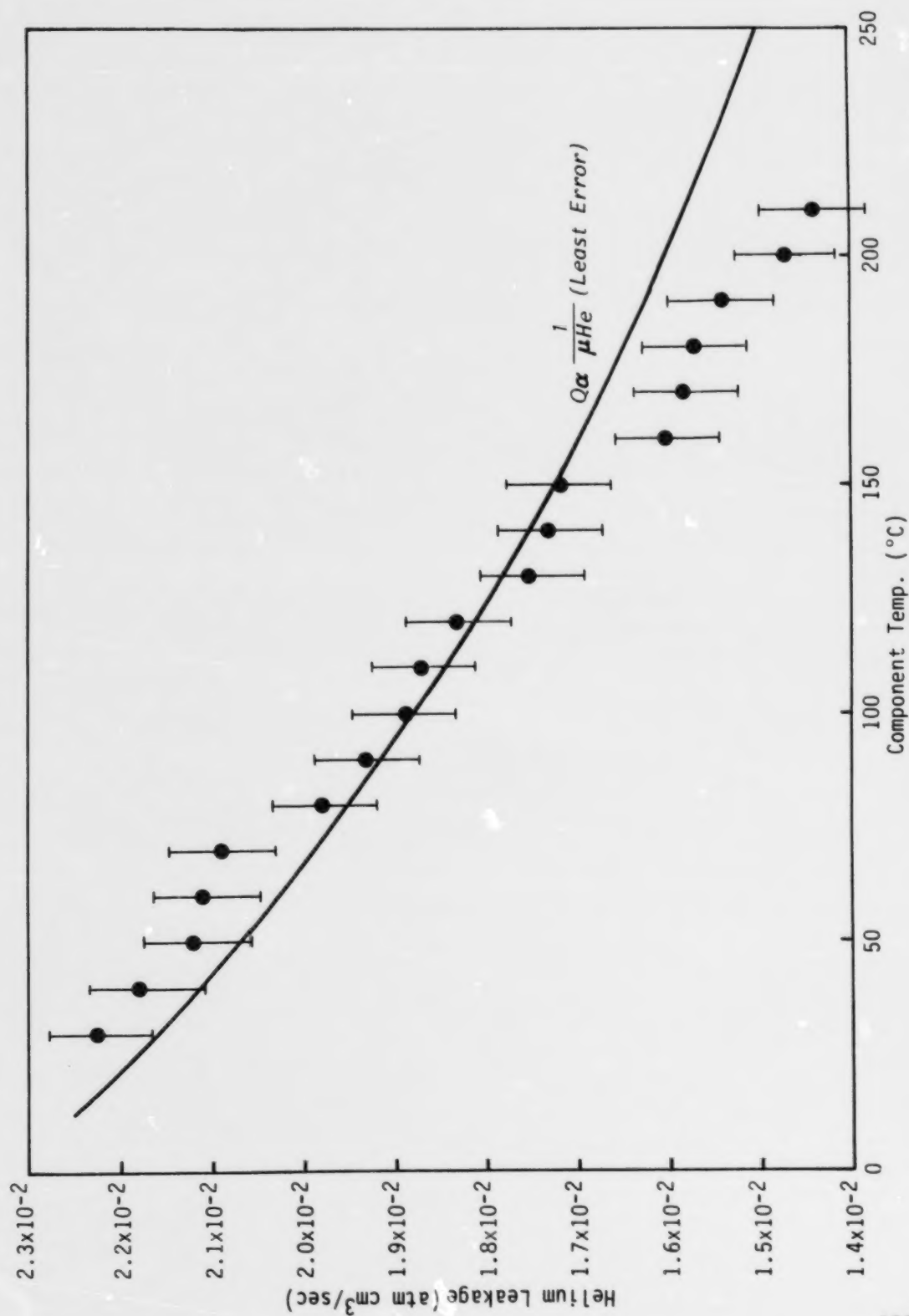


FIGURE 10
 HELIUM LEAKAGE VS COMPONENT TEMP.
 -3/8" Gyrolok Needle Valve# 3812G6
 - ΔP Across Seat = 9.8 MPa

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87-311

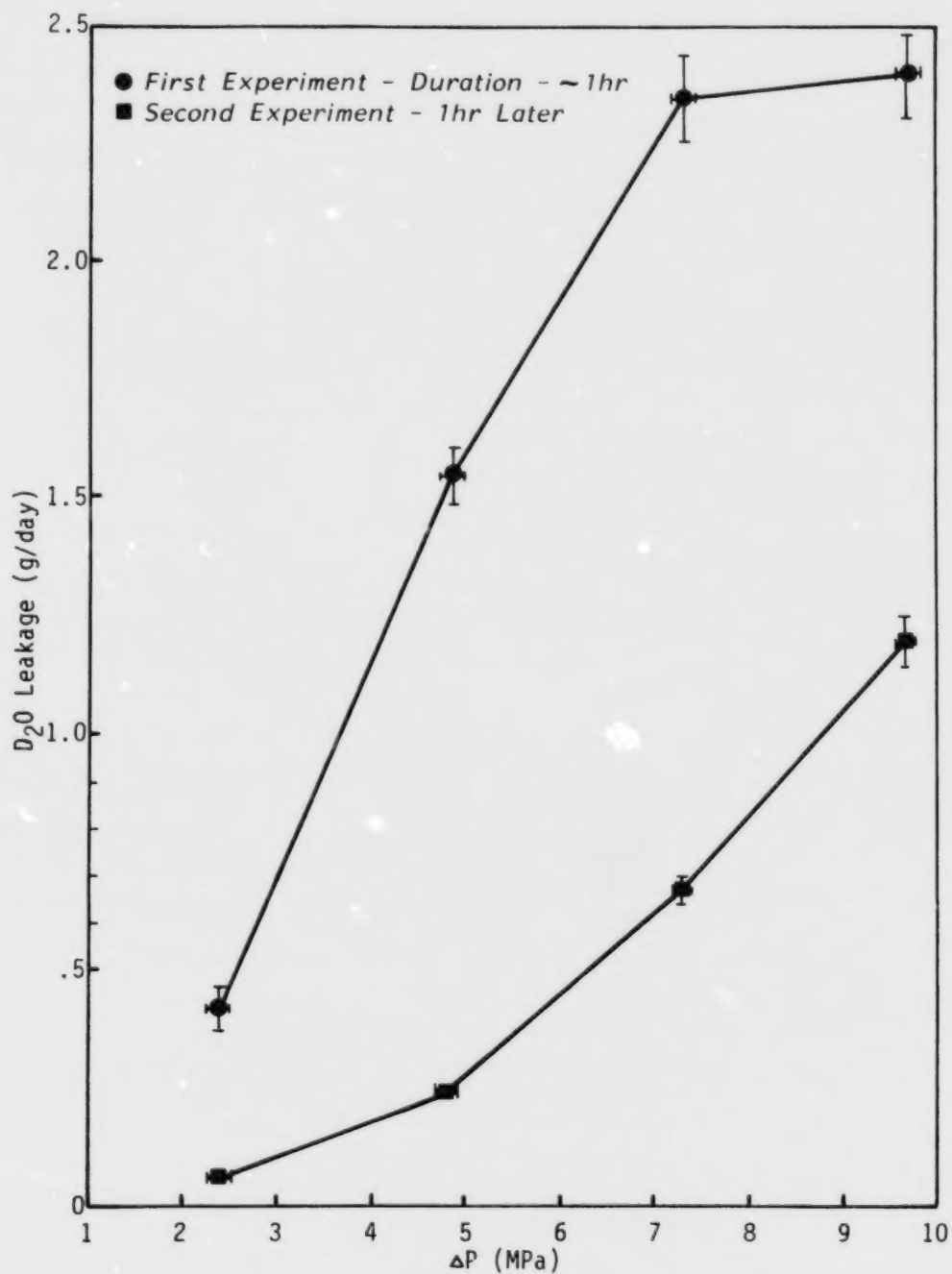


FIGURE 11
D₂O LEAKAGE VS ΔP
-Two Successive Series
-120°C

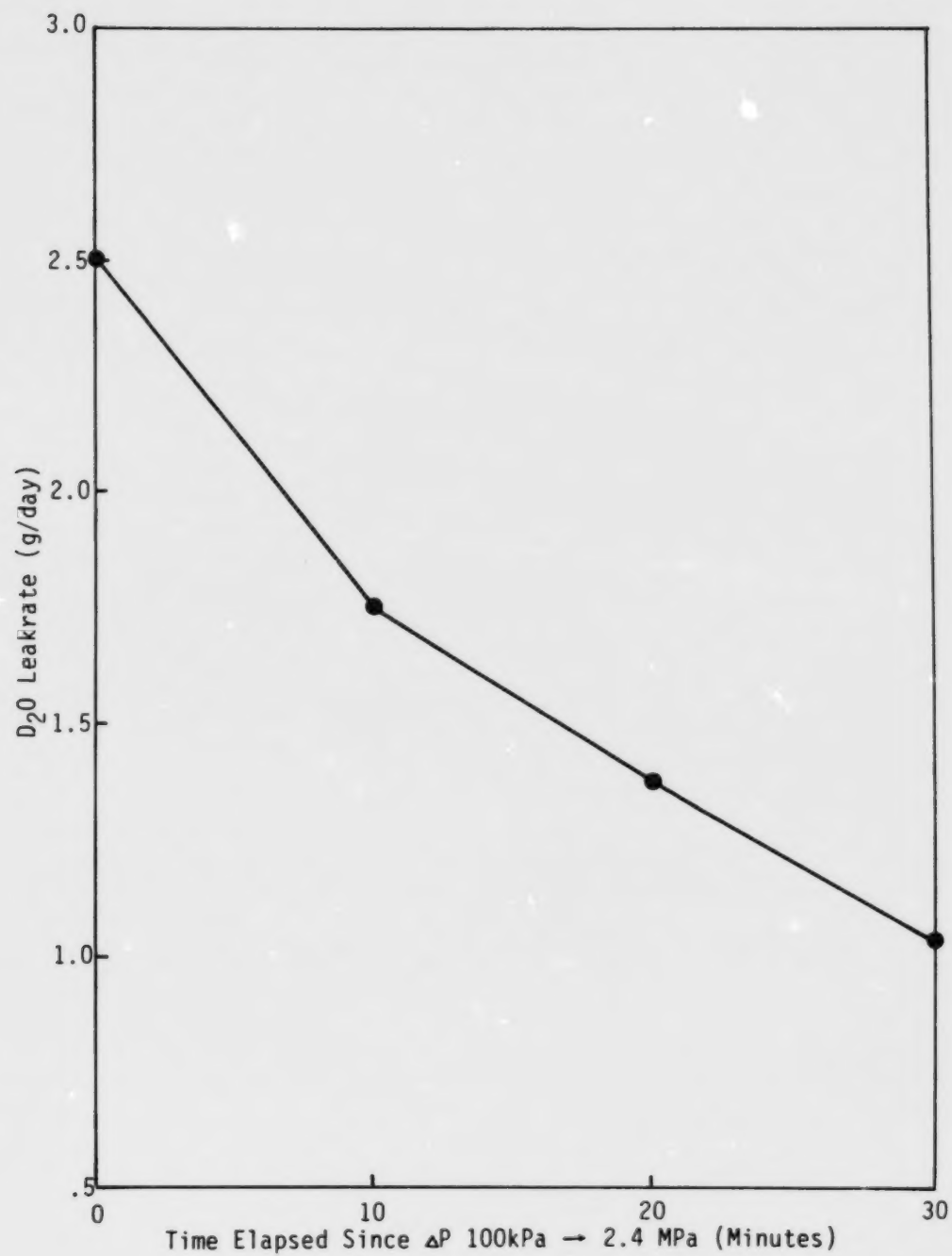


FIGURE 12
D₂O LEAKRATE VS TIME ELAPSED SINCE ΔP INCREASE

T = 120°C
P = 2.5 MPa (Up From 100 kPa @ T=0)

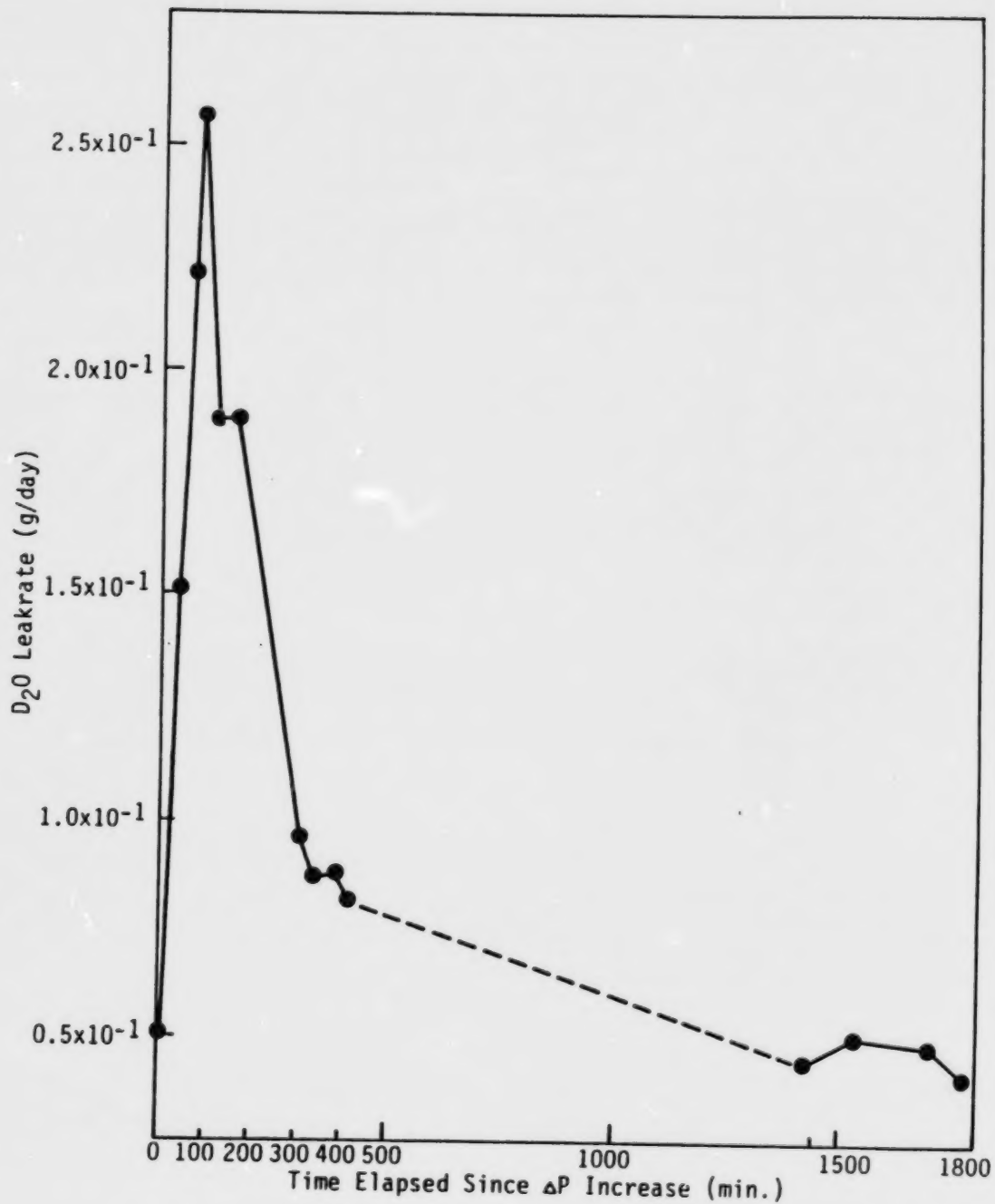


FIGURE 13
D₂O LEAKAGE VS TIME ELAPSED SINCE ΔP INCREASE FROM 100kPa TO 2.4MPa

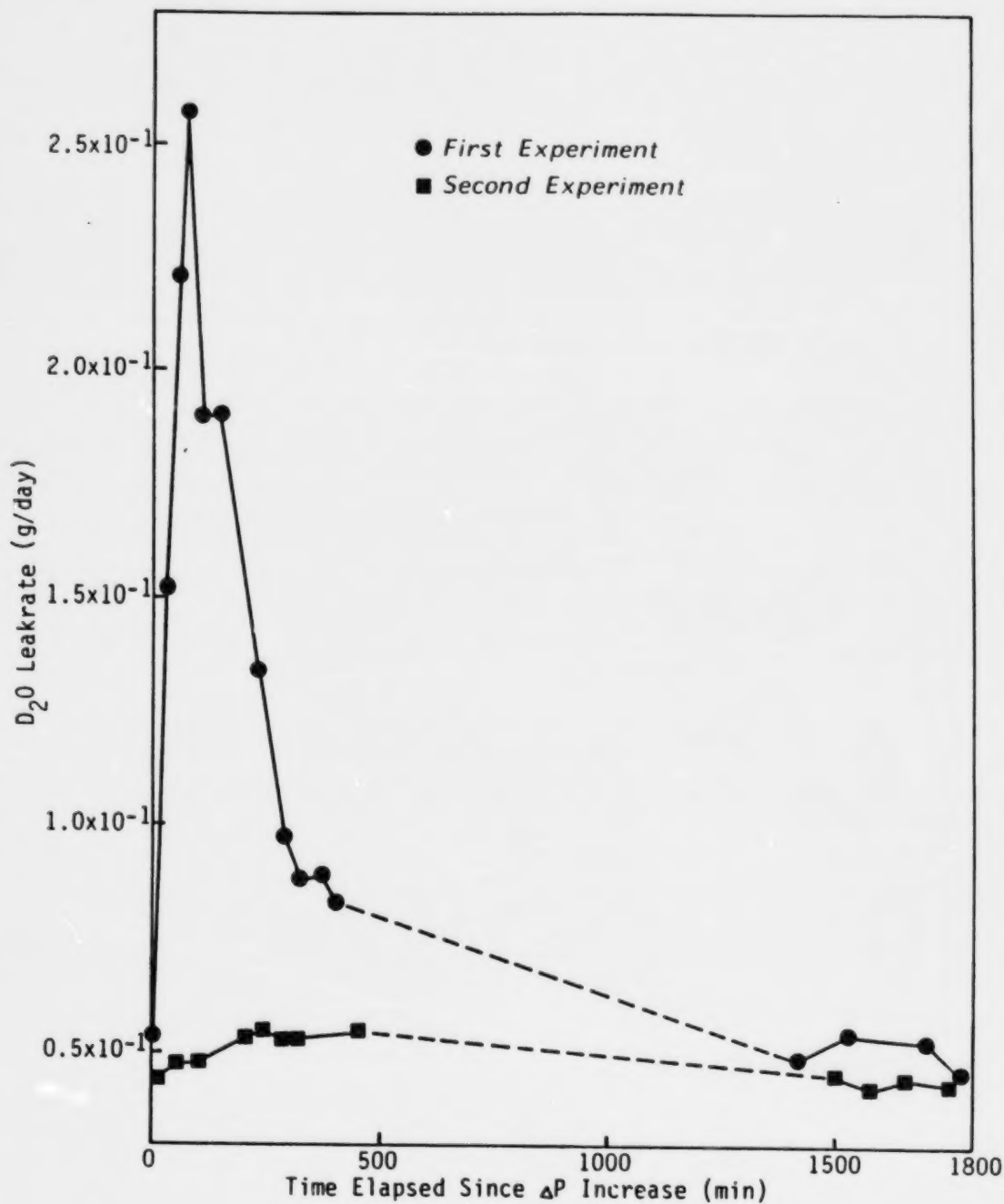


FIGURE 14
D₂O LEAKAGE VS TIME ELAPSED SINCE
 ΔP INCREASE FROM 100kPa TO 2.4MPa
-Two Successive Experiments

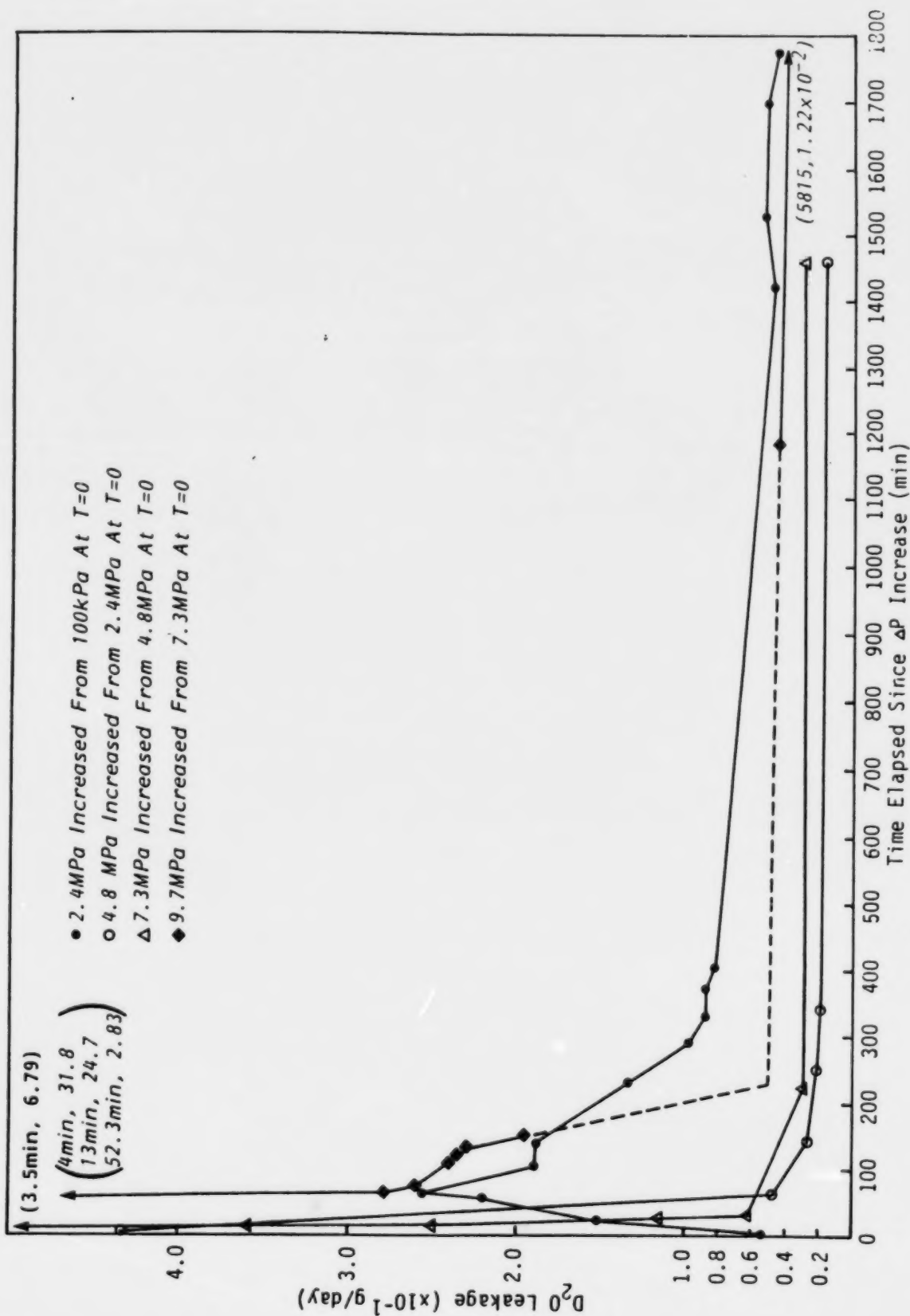


FIGURE 15

D_2O LEAKRATE VS TIME ELAPSED
 SINCE PRESSURE INCREASE

- 3/8 gyrolok valve
 - Temp=120°C

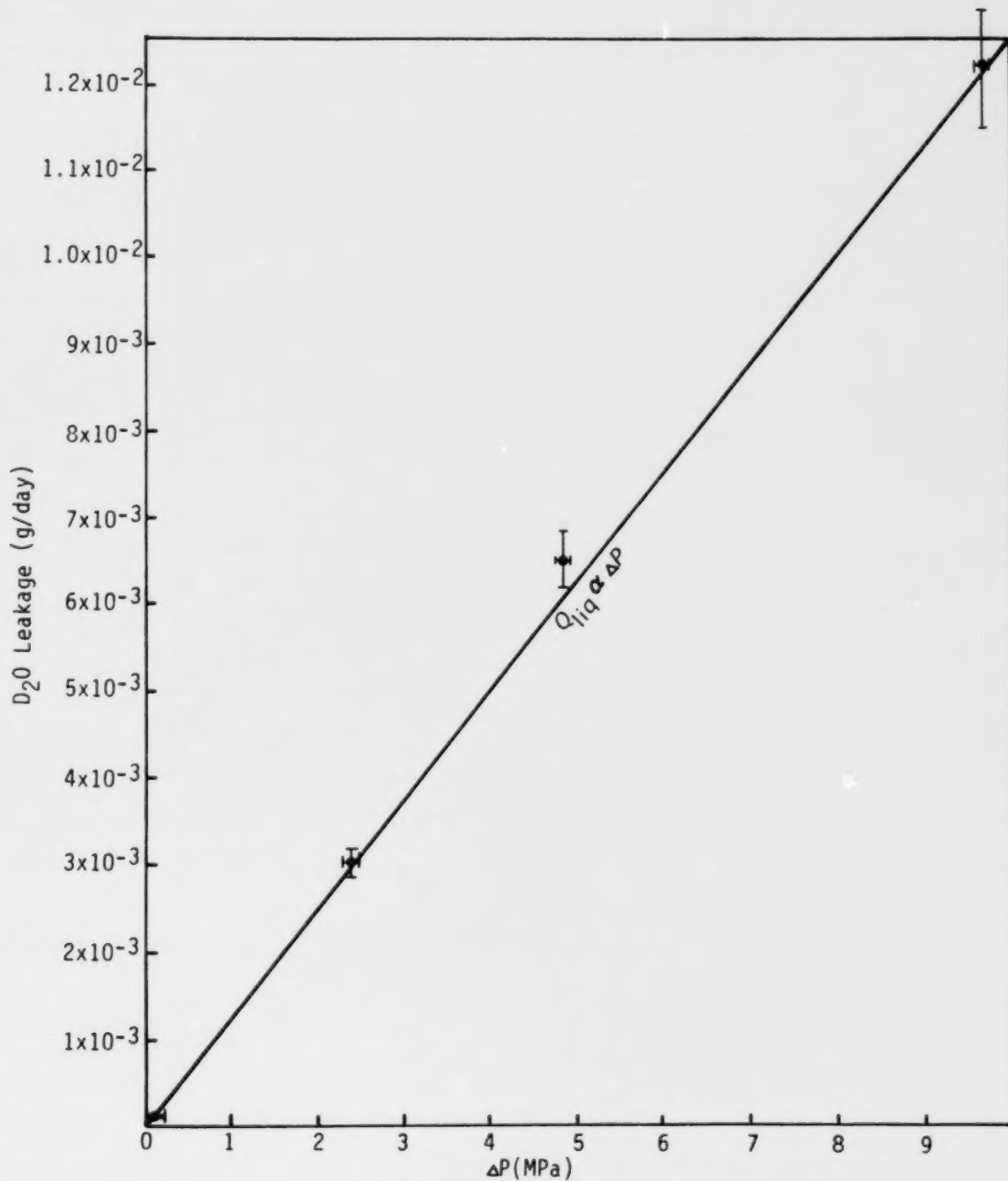
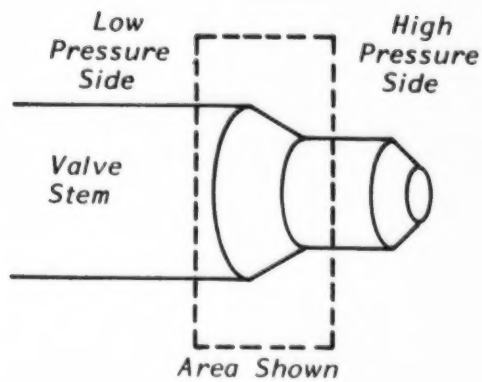
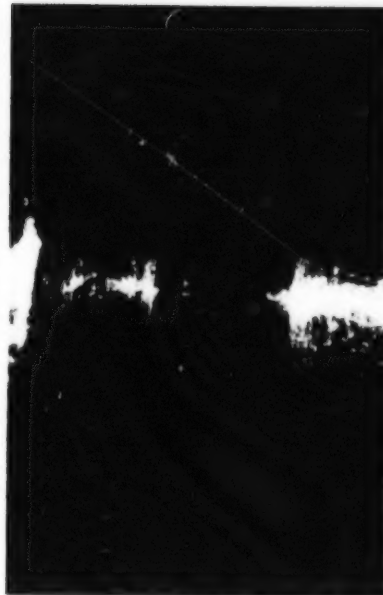


FIGURE 16
D₂O LEAKAGE VS ΔP

-Measured With Decreasing Pressure
after Leakrate had Stabilized at 9.7MPa



"Virgin Stem"



Stem After Experiments

One View

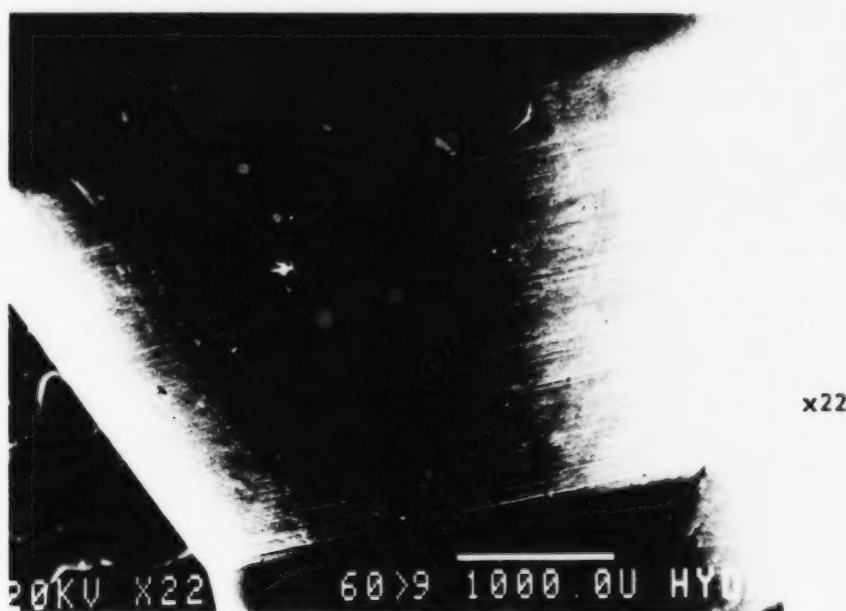


Rotated ~ 180°

FIGURE 17
VIEWS OF THE VALVE STEM IN THE AREA OF VALVE BODY CONTACT
-"Before" And "After"



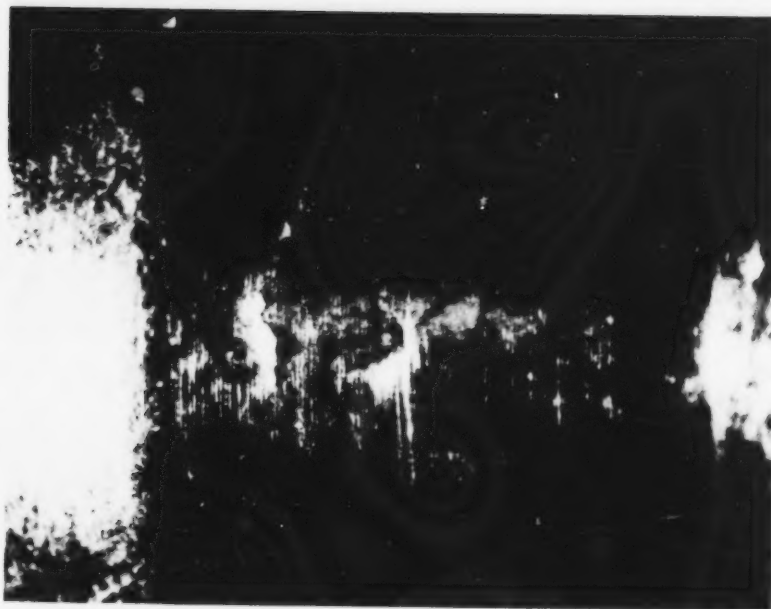
(a)



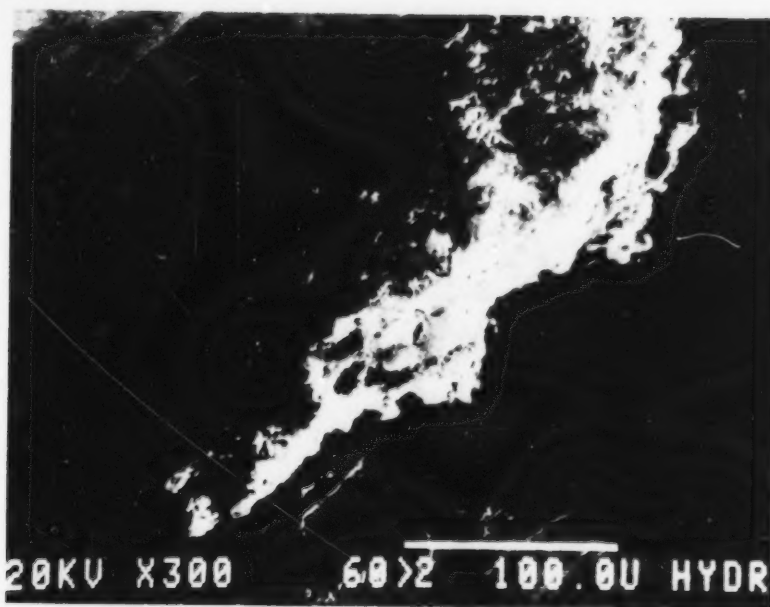
(b)

FIGURE 18

(a) SEM MICROGRAPH SHOWING THE CIRCUMFERENTIAL RING OF RESIDUE
 (b) LOWER PHOTO SHOWS A "VIRGIN" STEM



x40
(Optical)



x300
(SEM)

FIGURE 19
OPTICAL AND SEM MICROGRAPHS SHOWING ONE
OF TWO OBSERVED "WHITISH DEPOSITS"

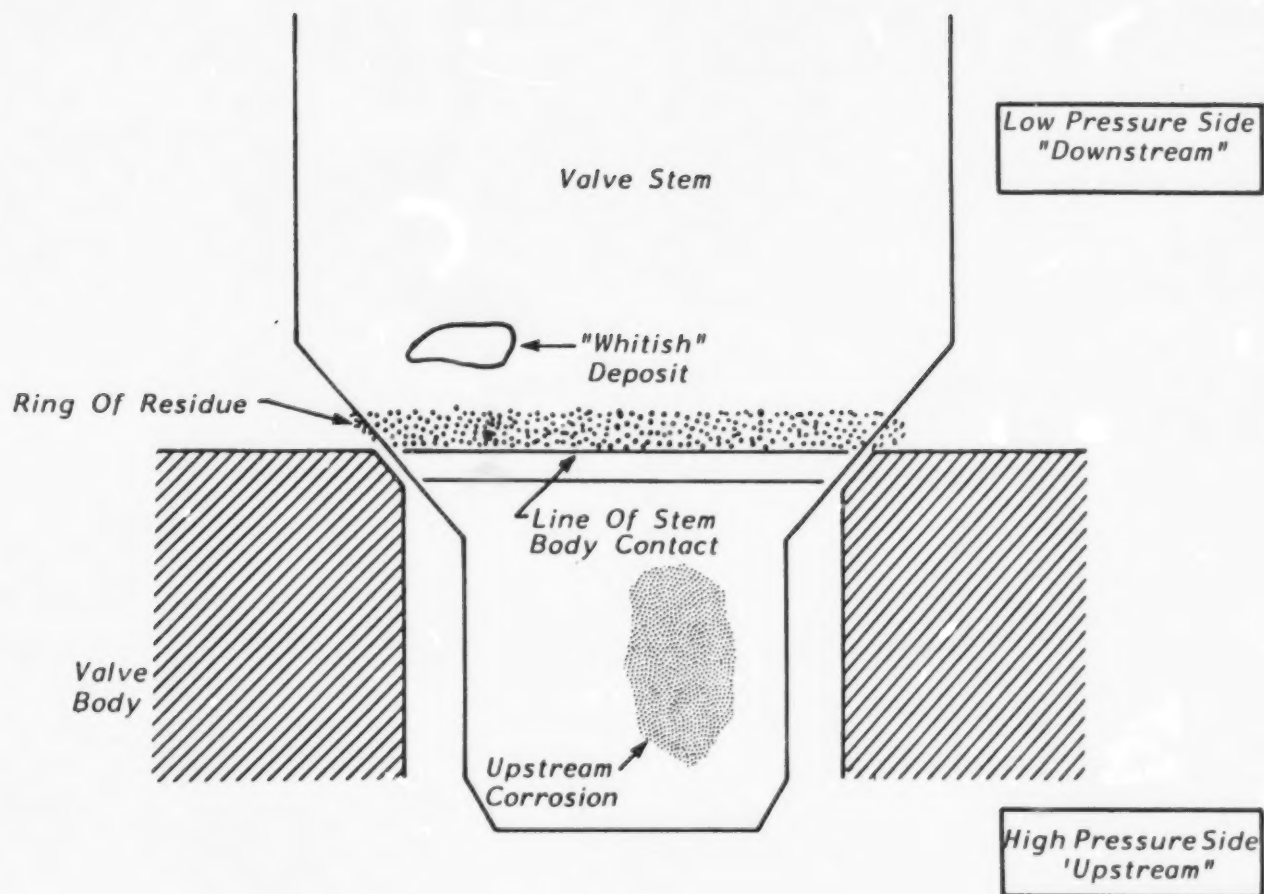
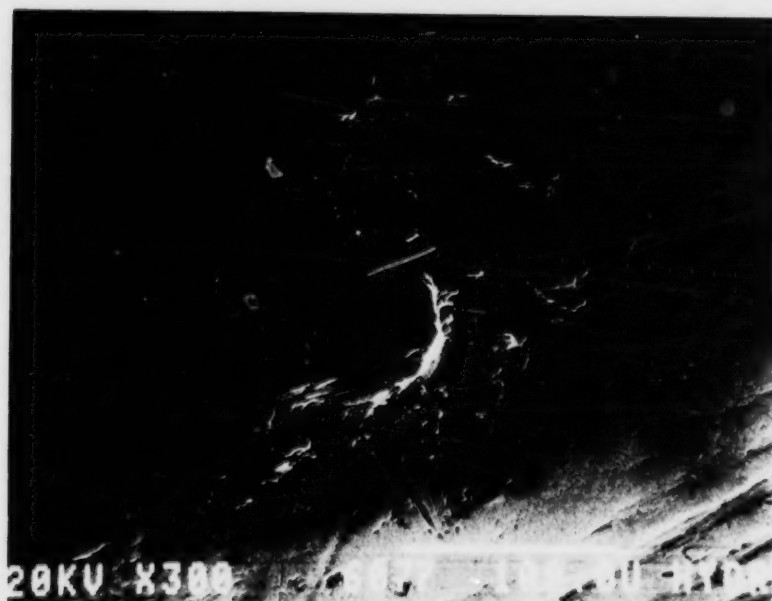


FIGURE 20
VALVE STEM AND BODY CONFIGURATION
SHOWING LOCATION OF OBSERVED FEATURES



magx300

FIGURE 21
A POSSIBLE LEAKAGE PATH ON THE STEM-SEAT
INTERFACE LINE OF THE VALVE STEM

Element	316SS	Percentage Composition By Weight (X-Ray Analysis)		
		Virgin Stem	"Rusty" Area	White Deposit
Si	1.00 Max	0.443	.374	0.700
S*	0.030 Max	0.958	.841	27.833
Cl			.510	12.514
K				0.659
Cr	16-18	20.906	20.116	1.359
Mn	2.00 Max			50.718
Fe	Remainder	66.597	67.987	4.109
Ni	10-14	11.096	10.172	1.102
Cu				0.444
Zn				0.563
C	0.08 Max			
P	0.045 Max			
Mo*	2-3			

*Note: Mo And S Are Hard To Separate Using This Technique.
The Analyzed "S" Percentage Should Be Taken To Include Mo.

FIGURE 22
PERCENTAGE COMPOSITION OF AREAS OF INTEREST
OF THE VALVE STEM (BY X-RAY ANALYSIS)

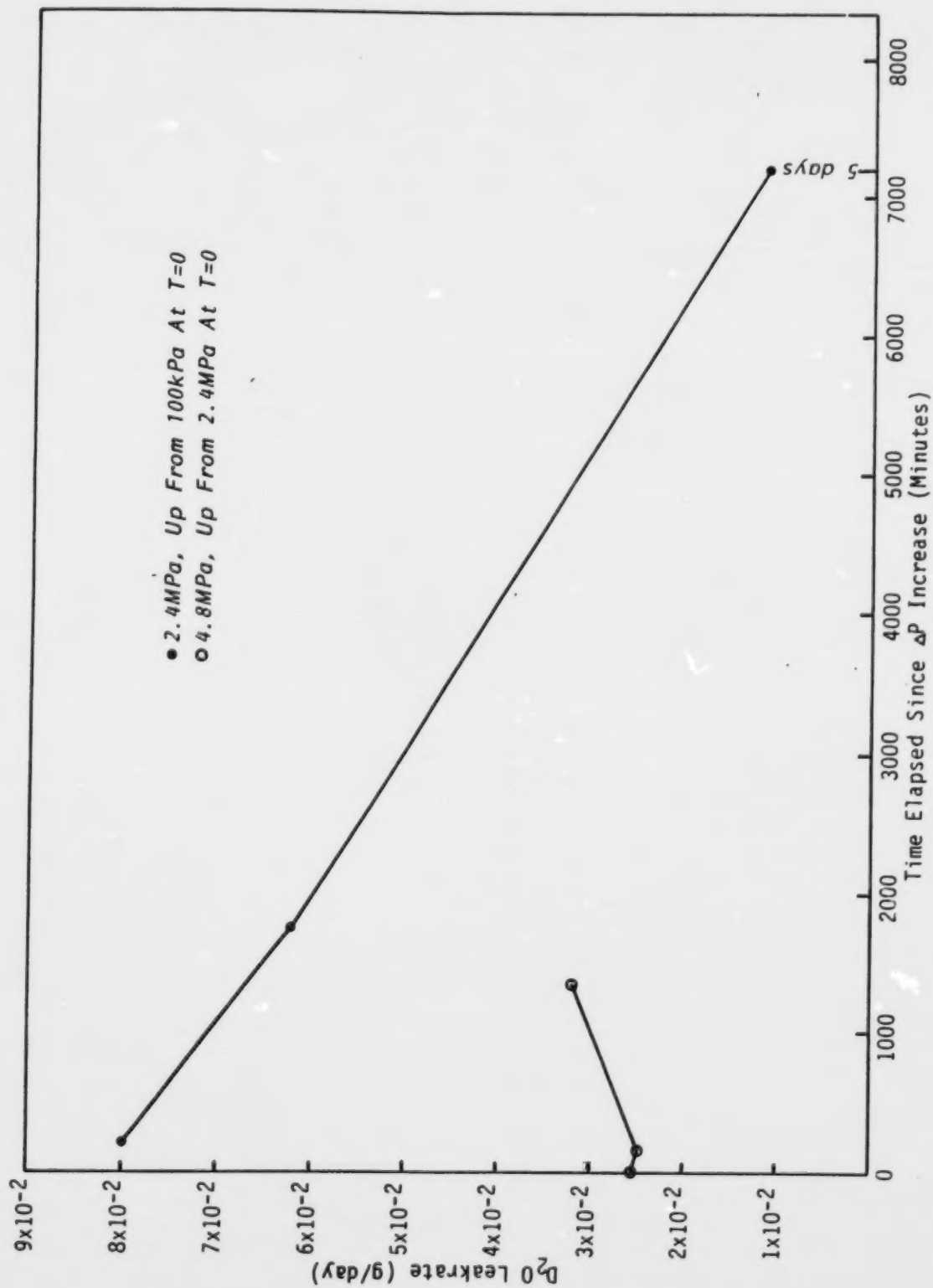


FIGURE 23
 D₂O LEAKRATE VS TIME ELAPSED SINCE ΔP INCREASE
 - Second 3/8 gyrolok valve
 - Temp=120°C

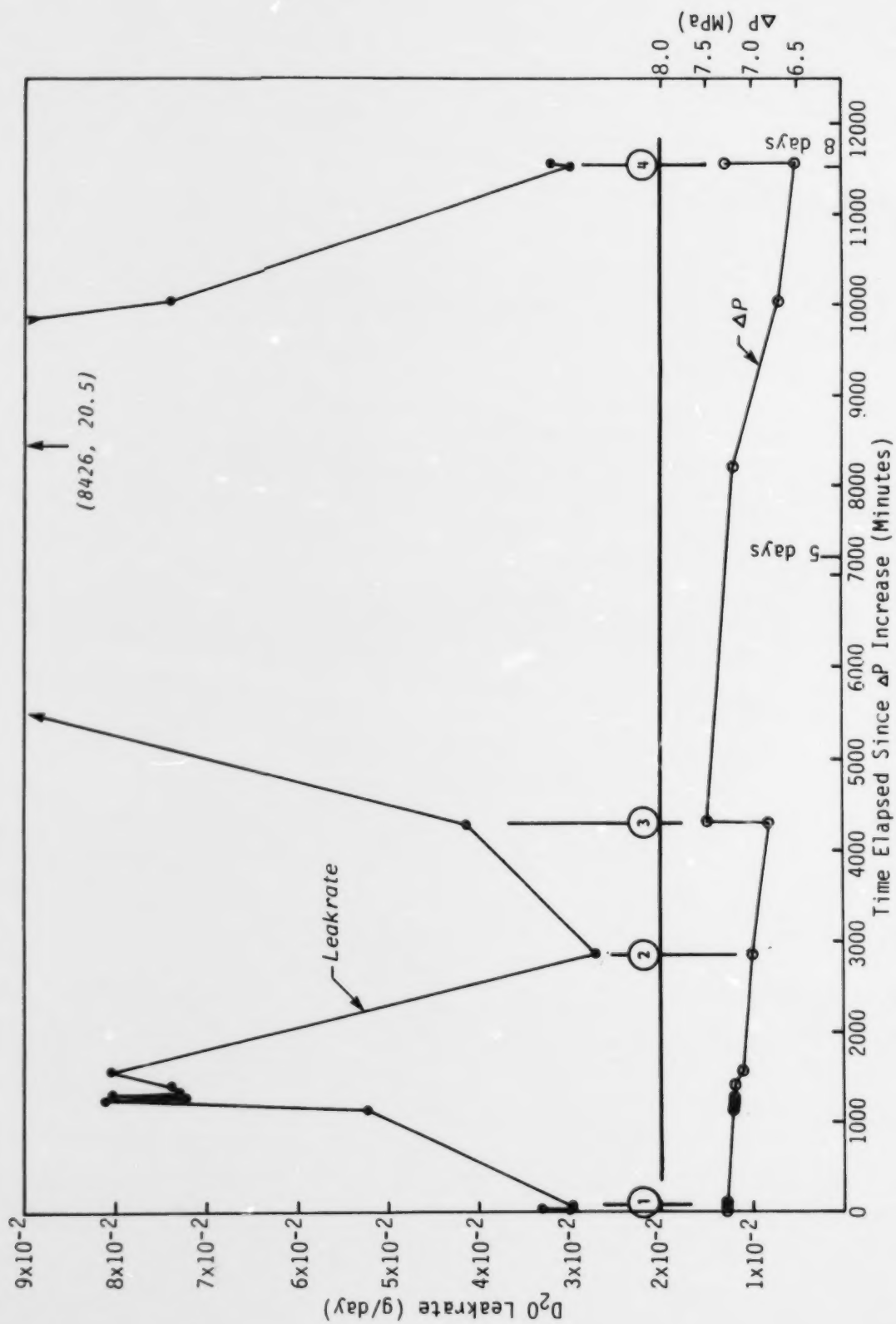


FIGURE 24
 D_2O LEAKRATE VS TIME ELAPSED SINCE ΔP INCREASE
 - Second 3/8 gyrolok valve
 - $T=120^\circ C$